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# UNITED STATES PATENT OFFICE

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PROCESS OF PRODUCING RADIO-ACTIVE ELEMENTS

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9 Claims. (Cl. 204-31)

This invention concerns methods and apparatus for the generation of radio-active bodies.

According to one feature of my invention, radio-active elements may be produced from nat-

- 5 ural elements by bombarding a natural element or compounds of natural elements with neutrons produced in various ways, more particularly, by subjecting the natural elements to neutrons emanating from a target containing lithium, which
- 10 target is subjected to a bombardment with fast deuterons. Another feature of the invention is directed to the production of radio-active elements from natural elements by exposing the natural elements to an irradiation with neutrons
- 15 which are liberated from certain elements under the action of X-rays. Another feature of the invention is directed to chemically concentrating radio-active elements produced from natural elements if the radio-active element is isotopic
  20 with the natural element from which it is pro-
- duced.

Other features of the invention will appear in the following detailed description referring to the drawings, and will be more particularly pointed 25 out in the claims.

In the drawings,

Figure 1 represents a sectional elevation of an apparatus for carrying out the invention,

Figure 2 shows a more constructional lay-out 30 of the apparatus of Figure 1,

Figure 3 shows the circuit arrangements for further modified apparatus and,

Figure 4 is a sectional view of apparatus intended to co-operate with that shown in Figure 3.

- 35 Referring first to Figure 1 of the drawings, II is an electrical discharge tube adapted to project a beam 12 of fast deuterons. The tube II is filled with deuterium and an anode A and cathode B are provided for connection to a source
- 40 of high voltage. The deuterons are thus projected at high speed and pass through the cathode B. The deuterons fall on a substance 13 in a sealed container 13A. The substance 13 consists, for instance, of lithium. The collision of the fast
- 45 diplogen ions with the substance 13 causes transmutation, i. e. a nuclear reaction of the deuteron with an atom of the target. The substance 13 is surrounded by a thick layer 14 containing the element which it is desired to transmute into a
- 30 radio-active element. In order to have a high efficiency, the thickness of the layer 14 has to be sufficiently great, compared with the mean free path of the neutron, to prevent escape of any of the neutrons.

55 Figure 2 shows in more detail the electrical dis-

charge tube 11 referred to in Figure 1. The tube essentially consists of a main portion 16 serving to accelerate the deuterons and an auxiliary tube **IIT** for initiating the flow. **IIA** is the anode and 15 the cathode of the auxiliary tube, deuterium 5 being admitted thereto through the inlet 13B and being pumped away through the outlet 14A. The flow initiated by the auxiliary tube is accelerated by passage through the main tube 16 which is maintained exhausted by suction outlets 141 and 10  $14^2$ , and which has a high potential gradient. there being a million volt potential difference between the ends of the tube. The accelerated deuterons emerge through the neck  $14^3$ , of the tube 16 and collide with the substance 13 as de- 15 scribed with reference to Figure 1 of the drawings.

If the substance 13 is a light element for instance lithium, then the bombardment by the accelerated deuterons results in emission of un- 20 charged particles of mass of the order of magnitude of the mass of a proton. Such uncharged nuclei i. e. neutrons, penetrate even substances containing the heavier elements without ionisation losses, and will cause the formation of radio- 25 active substances in the layer 14 exposed to them. It is to be noted that by the method so far described, the ionisation losses suffered by the deuterium nuclei are comparatively small in light elements and also that the substance to be made 30 radio-active is irradiated with neutrons i. e. uncharged nuclei, which pass through even heavy elements without ionising them. The substance 14 exposed for treatment by the neutron radiation may be in the form of an organic compound 35 for the purpose of carrying out separation of the generated radio-active element, as described more fully hereinafter.

Neutron radiation may also be produced by the action of X-rays upon an element having a dis- 40 sociable neutron at the prevailing voltage, and apparatus for carrying out this process will now be described with reference to Figure 3 of the drawings.

In Figure 3, 1 is the primary of a transformer, 45 the secondary 2 of which is connected to the junctions 3 and 4. The junction 3 is connected to the cathode 8 of the rectifier tube 5 and to the anode 7 of the rectifier tube 6. The junction 4 is connected to the cathode 9 of the rectifier 50 tube 10 and to the anode 11 of the rectifier tube 12. The cathodes 13 and 14 are connected to each other and to earth. The anodes 15 and 16 are connected at 17, and fom this point are connected to the pole 18 of the impulse generator 20, 55 the pole 19 of which is connected to earth. The impulse generator 20 is built of condensers 21, resistances 22 and spark-gap devices 23. The impulse generator and rectifying unit

- s shortly described above, are known components adapted to give an extremely high voltage for a fraction of a second. With such a system volt-ages up to 3 million volts have been obtained. The negative side of the impulse generator is con-
- 10 nected to a spark gap device 25, which in turn is connected with the cathode 26 of the discharge tube 24. The latter is built up from rings 24A of which only a few are shown in the drawings. It will, however, be understood that the rings
- 15 are continuous to enclose a space which is exhausted through the outlet 24B. The anode 27 of the tube is connected to earth and is formed by a metallic window. A body of material 28 is arranged at the external side of the window 27.
- When the impulse generator operates to pro-21 duce discharge between the cathode 26 and anode 27 of the tube 24, fast electrons penetrate the anode 27 and impinge upon the body 28. The latter when formed of Bi or Pb or some other
- 25 heavy element, efficiently acts as an anti-cathode and hard X-rays are produced. In Figure 4 of the drawings there is shown the

lower portion of the discharge tube 24 with a device therebeneath for utilising the hard X-rays

- so capable of being produced with the aid of the fast electrons emerging through the anode 27 of the tube 24. The device consists of a block 34 of the element which is to be made radioactive, a block 32 of an element with a dissociable
- 35 neutron, being located therein. An aperture is formed in both the blocks 32 and 34 to allow entry of the cathode rays from the tube 24 above. The blocks 32 and 34 are also arranged to accommodate a wheel 30 and axle 35. The wheel 30
- 40 at its periphery carries a covering of tungsten or lead 31. The covering 31 acts as an anticathode and is cooled with water introduced along the bearing for the axle 35. The block 34 may be in the form of a cube having a length of
- 45 side of 50 cm., whilst the block 32 can also be of cube form with a side of 25 cm. For the sake of example the block 34 may be formed of iodine or arsenic or other material which lends itself to being made radio-active. The block 32 may
- 50 be of metallic beryllium. In order that an isotopic separation as described hereinafter may be performed after irradiation the material of the block 34 may be in the form of an organic compound. A voltage of 3 million volts may be used
- 55 for the discharge tube and in operation the wheel 30 is rotated so that electrons passing through the anode 27 of the tube 24 hit the rotating anticathode covering 31. When the fast electrons strike the anti-cathode, hard X-rays are pro-60 duced which penetrate the beryllium block 32
- and cause neutrons to be released therefrom, which neutrons then act upon the block 34.

It may be that fast electrons and hard X-rays have a similar effect upon beryllium and one may 65 therefore contemplate the making of the covering 31 of the wheel 30 from beryllium, the beryllium block 32 then being dispensed with, so that the neutrons released directly from the beryllium anti-cathode may enter and act upon the block 70 34.

It is found that when various elements are irradiated with neutrons by the process described above, practically all elements which become radio-active transmute into their own radio-

75 active isotopes, and it becomes difficult to sepa-

rate these radio-active isotopes from the remaining portion of the element unaffected. In order to achieve separation of the radio-active element from the non-radio-active part thereof the following process may be adopted. This process is based on the fact that if a compound of an element is irradiated by neutrons, and if an atom of the element transmutes into the radio-active isotope, then this atom is freed from the compound. In accordance with the process, a com- 10 pound of the element it is desired to make radioactive is chosen such that the freed radio-active isotope of the element will not interchange with the combined atoms of the element within the compound, whereby the freed isotope may be 15 chemically separated from the irradiated compound. Very often the element whose radioactive isotope is to be isolated, can be conveniently irradiated in the form of a compound in which it is bound to carbon. Thus in the case 20 of iodine compounds such as iodoform or ethyl iodide, the radio-active iodine isotope may be chemically separated from the original jodine compound in the form of free iodine. In order to protect the radio-active iodine isotope a small 25 amount of normal iodine may be dissolved in the organic iodine compound before irradiation or after irradiation but before separation.

What I claim and desire to secure by Letters Patent of the United States is:

1. The method of producing a radio-active element from a natural element by causing fast deuterons to impinge on a target containing lithium, and exposing a layer of the natural element to be transformed into a radio-active ele- 33 ment to the neutron radiation emitted by the said target.

2. The method of producing from a natural element a concentrate of a radio-active element which is isotopic with the said natural element, 40 which comprises subjecting a compound of said natural element to an irradiation which will transform some of said natural element into a radio-active isotope of said natural element, said compound of said natural element being one 4.5 which in the environment in which the irradiation is being carried out does not interchange atoms of said natural element bound in the compound with atoms of said natural element or its isotopes outside the compound, and separating, 50 after irradiation, from the compound said natural element and its isotopes which are outside the compound.

3. The method of producing from a natural element a concentrate of a radio-active element 55 which is isotopic with said natural element, which comprises subjecting a compound of said natural element to irradiation with neutrons which will transform some of said natural element into a radio-active isotope of said natural ele- co ment, said compound of said natural element being one which does not interchange in the environment in which the irradiation is carried out, atoms of said natural element bound in the compound with atoms of said natural element 63 or its isotopes outside the compound, and separating, after irradiation, from the compound said natural element and its isotopes which are outside the compound.

4. The method of producing from a natural 70 element a concentrate of a radio-active element which is isotopic with said natural element. which comprises irradiating with neutrons an organic compound of said natural element which will not interchange atoms of said natural ele- 75

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ment bound in the compound with atoms of said natural element or its isotopes outside the compound, and separating, after irradiation, from the compound said natural element and its isotopes outside the compound.

5. The method of producing from a natural element a concentrate of a radio-active element which is isotopic with said natural element, which comprises irradiating with neutrons a compound

- 10 which contains carbon, in which said natural element is bound to carbon and which compound will not interchange atoms of said natural element bound in the compound with atoms of said natural element or its isotopes outside the com-15 pound, and separating, after irradiation, from
- the compound said natural element and its isotopes outside the compound.
  - 6. The method of producing from a natural element a radio-active element which is isotopic
- 20 with the natural element comprising the steps of producing fast electrons, directing them toward a target adapted to produce X-rays under the impact of said electrons, exposing to the action of said X-rays an element of the class
- 25 consisting of beryllium and heavy hydrogen which produce neutron radiation under the action of said X-rays, and producing a radio-active element from a natural element by exposing the natural element to said neutron radiation.
- **30** 7. The method of producing from a natural element a radio-active element which is isotopic with the natural element comprising the steps of producing fast electrons having an energy of at least 3,000,000 volts, directing them toward a
- **35** target adapted to produce X-rays under the impact of said electrons, exposing to the action of

said X-rays an element of the class consisting of beryllium and heavy hydrogen from which neutrons are liberated by X-rays of 3,000,000volts energy, and producing a radio-active element from a natural element by exposing the 5 natural element to said neutron radiation.

8. The method of producing from a natural element a radio-active element which is isotopic with the natural element comprising the steps of producing fast electrons, directing them toward a target adapted to produce X-rays under the impact of said electrons, exposing beryllium to the action of said X-rays to produce neutron radiation, and producing a radio-active element from a natural element by exposing the natural element to said neutron radiation.

9. The method of producing from a natural element a radio-active element which is isotopic with said natural element comprising the steps of producing fast electrons, directing them to-20 ward a target adapted to produce X-rays under the impact of said electrons, exposing to the action of said X-rays an element of the class consisting of beryllium and heavy hydrogen which produce neutron radiation under the action of 25 said X-rays, and irradiating by said neutron radiation a compound of said natural element which in the environment in which said irradiation is carried out will not interchange atoms of said natural element bound in the compound with 30 atoms of said natural element or its isotopes outside the compound, and separating, after irradiation from the compound said natural element and its isotopes outside the compound.

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The invention concerns a process and apparatus for 1 the transmutation of elements and makes it possible to 2 bring about such a transmutation on a large scale. 3 The production of power by means of the heat generated in the 4 process and the production of radio active elements on a 5 large scale are thus made possible by the invention. 6 The 7 invention teaches that it is possible to produce a nuclear chain reaction and to maintain the stationary conditions in 8 such chain reactions. It also teaches that it is possible 9 to have explosive bodies in which an explosion is brought 10 about at will by a sudden change in the distribution of 11 matter. 12

13 According to the invention such a chain reaction may be maintained for instance by maintaining an initial 14 radiation of neutrons by means of one of the known methods 15 for the production of neutron radiations (for instance by 16 17 bombarding a lithium target with deuterons or by exposing beryllium to X-rays or gamma-rays) and letting these neu-18 of trons fall on a body/suitably chosen substance, shape and 19 dimensions and thereby obtaining an increase in the number 20 and energy of the neutrons which increase is brought about 21 by their interaction with the said body. Thus if the 22 proper conditions are observed the interaction of neutrons 23 with matter can lead to the liberation of further neutrons. 24 These newly liberated neutrons liberate again in their turn 25 further neutrons so that we have a chain reaction in which 26 a large number of neutrons are liberated, the total number 27 being limited only by the dimensions of the geometrical 28 arrangement. 29

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Such change is not only possible in the presence of a meta 1 stable element. A meta stable element is an element the 2 3 mass of which (packing fraction) is sufficiently high to allow its disintegration into its parts and a liberation of 4 5 energy. Elements like uranium and thorium are exemples of such elements. It is possible to measure the packing 6 7 fraction of the elements by means of the mass spectograph 8 and thereby determine whether an element is meta stable or 9 not in the sense in which the word is used in this application. If, for instance, the values which were generally 10 11 accepted in 1934 for helium and beryllium had been correct 12 (and we know today that they were false, at least in their 13 relation to each other), we have been correct in concluding 14 from these values which were accepted in 1934 that beryllium 15 is a meta stable element and can be disintegrated into parts, 16 with a liberation of energy, one of the parts set free in 17 its disintegration being a neutron. If such an element which 18 is meta stable but the disintegration of which is inhibited 19 is exposed to neutrons, we may have a reaction in which a 20 neutron disappears and more than one neutron is emitted. 21 The additional neutrons would together with the number of 22 the original neutrons continue to interact with the meta 23 stable element thereby forming the links of a chain reaction.

In order to be able to utilize a nuclear reaction in which an excess number of neutrons is liberated by neutrons for the maintenance of a chain reaction, it is not sufficient to measure the cross-section and other constants of the reaction, but it is also necessary to be aware of the laws which govern the neutron output of such reactions in function of the geometrical conditions. Once the general laws, the

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type of behavior is known, the exact dimensions can be
 easily determined in each particular case by actually
 measuring the neutron output. Such experimental adjustments
 can, however, only be made if the general type of behavior
 is known.

In the following we shall by way of example demon-strate certain general features of such a neutron chain reaction in a special case. In this special case the carrier of the chain reaction forms a spherical layer as illustrated in Fig. 1. In Fig. 1, 1 is a spherical layer which contains a carrier of the chain reaction, 2 is a neutron source which has fixed constant output of neutrons,  $\underline{r}_1$  is the inner radius of the spherical layer, and  $\underline{r}_2$  is the outer radius of the spherical layer. In order to be able to treat the problem as a diffusion problem, we choose  $\underline{r}_1$  very much larger than the mean free path  $\underline{a}$  of the neu-trons produced in the chain reaction in the substance of 

the chain reaction layer. The neutrons liberated in the 1 chain reaction layer will have to make a number of collisions 2 with the nuclei which compose the chain reaction layer, and 3 we designate the number of collisions necessary for causing 4 transmutations in the carrier of the chain reaction layer 5 which lead on the average to the liberation of one addi-6 tional neutron with f. We further assume that f is a large 7 number so that we may have conditions in which the well 8 known equations of diffusion can be applied. With these 9 assumptions the density s of the neutron will with good 10 approximation be given within the chain reaction layer as 11 a function of the radius  $\underline{r}$  by the following equation: 12 (1)  $D \frac{d^2(r_5)}{dr^2} + A(r_5) = 0$ 13 D is a diffusion constant which is given by 14  $D = \frac{av}{31}$ 15

16  $\checkmark$  is the mean velocity of the neutrons; <u>a</u> the mean free path 17 for scattering collisions within the chain reaction layer; 18 and <u>f</u> a number which says how many scattering collisions 19 a neutron has to make in the chain reaction layer in order 20 to produce on the average an additional neutron.

<u>A</u> stands for the number of neutrons produced per c.c. of chain reaction layer in a second and its value is given by

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 $A = \frac{v}{aF}$  so that  $\sqrt{\frac{D}{A}} = \frac{aVF}{V3}$ 

For a given value of  $\underline{\mathbf{r}}_1$  there is a certain value of  $\underline{\mathbf{r}}_2$ and accordingly a certain value of  $\underline{\mathbf{r}}_2 - \underline{\mathbf{r}}_1$  for which the number of neutrons per second diffusing out of the spherical layer into space becomes infinite for a finite neutron production of the source. This value of  $(\underline{\mathbf{r}}_2 - \underline{\mathbf{r}}_1)$  we may call the critical thickness of the chain reaction layer. If the

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thickness of the chain reaction layer is smaller than but 1 very close to the critical thickness, the neutron output 2 is very much larger than the neutron input and we may have 3 1000 or more times as many neutrons emerging from the chain 4 reaction layer as the neutron input of the neutron source 2. 5 The value of the critical thickness is a function of  $r_1$ 6 and of the boundary conditions for  $\underline{r} = \underline{r}_1$  and  $\underline{r} = \underline{r}_2$ . If 7 the outer surface  $(\underline{r} = \underline{r}_2)$  of the spherical layer were to 8 stand free in space, the density s would be 0 for the outer 9 surface, and if there is no absorption of neutrons in the 10 hollow sphere containing the neutron source, i.e. if the 11 number of neutrons produced by the neutron source is equal 12 to the number of neutrons diffusing outwards from the 13 sphere  $\underline{r} = \underline{r}_1$ , we obtain very large values of  $\underline{r}_1 (\underline{r}_1 / / D_A)$ 14 for the value of the critical thickness 15

17 The critical thickness for these conditions we shall call 18 further below the standard critical thickness of the sub-19 stance which composes the chain reaction layer.

16

If the outer surface is covered by some material, 20 for instance if the transmutation layer is covered by lead 21 or immersed into water, the critical thickness is less than 22 the standard critical thickness. This us due to the back 23 scattering by lead or water, and, in the case of water, 24 the fact that the neutrons are slowed down by water and their 25 mean free path is thereby reduced plays an important role. 26 If the neutrons are allowed to escape out of the 27

hollow sphere containing the neutron source in the interior of the spherical layer or if they are absorbed within this hollow sphere, the critical thickness is increased.

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If all the neutrons were captured in the hollow sphere within  $\underline{r} = \underline{r}_1$ , the critical thickness would become independent of the value of  $\underline{r}_1$  and have a value exactly double the normal critical thickness provided that the neutron density  $\underline{s}$  remains 0 at the outer surface  $(\underline{r} = \underline{r}_2)$ .

Obviously the above given diffusion equation holds 6 only for stationary solutions, that is for solutions where 7 the neutron density  $\underline{s}$  is a function of  $\underline{r}$  only and does 8 not vary with time. Not for all boundary conditions will 9 such stationary solutions exist. If we gradually increase 10 the thickness of the critical layer we reach for any given 11 12 boundary conditions a thickness at which the neutron outflow becomes infinite for finite neutron production of the 13 neutron source. This thickness is a critical thickness 14 15 of the arrangement. It can be calculated for every case in 16 the following way from given boundary conditions.

17 The solution of the above given diffusion equation18 has the form of

19 (3)  $Jr = C_{sin}(V = h) + C_2 coe(V = h)$ 

For the boundary condition  $\underline{s} = 0$  for  $\underline{r} = \underline{r}_2$  the solution 21 takes the form of

- (4) sr= Csin[Cr2-2) V = ]
- 23

22

24 (5)

or

D= C sin [C+2-D V 7/A]

If there is no absorption of neutrons in the hollow sphere around the neutron source and if the neutron source produces a fixed number N of neutrons per second, the number of neutrons diffusing from the sphere  $\underline{r} = \underline{r}_1$ into the chain reaction layer must be equal to N. On the other hand, the number of neutrons thus diffusing through any sphere is given by  $4\pi r^2 \int \frac{ds}{dr}$ 

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and it is therefore also  $N = 4\pi \tau^2 D \frac{\mathbf{D}}{ar} + 4\pi \tau^2 \frac{\mathbf{D}}{ar}$ 

For a value of  $\underline{\mathbf{r}} = \underline{\mathbf{r}}$  for which  $\underbrace{df}_{dT}$  becomes 0, the ratio of the number of neutrons diffusing through the outer surface  $\underline{\mathbf{r}} = \underline{\mathbf{r}}_2$  to N becomes infinite. The corresponding which value of  $(\underline{\mathbf{r}}_{\mathbf{d}} - \underline{\mathbf{r}}_1)$  is therefore the critical thickness and value of  $(\underline{\mathbf{r}}_{\mathbf{d}} - \underline{\mathbf{r}}_1)$  is therefore the critical thickness and formation (5) what thus be calculated for the values of  $\underline{\mathbf{r}}_1$ . For very small values of  $\underline{\mathbf{r}}_1$  ( $\frac{1}{7} \ll \left\lfloor \frac{D}{A} \right\rfloor$ ) it obviously becomes twice the standard critical thickness  $\hat{\boldsymbol{L}} \cdot \boldsymbol{e} \cdot \mathcal{T} / \frac{D}{A}$ 

To take another example, if the boundary conditions are  $\underline{s} = 0$  both for  $\underline{r}_1$  and  $\underline{r}_2$ , then a glance at equation (5) shows that  $(\underline{r}_2 - \underline{r}_1) = \overline{n} / \frac{D}{A}$ , i.e. twice the critical thickness, quite independent of the value of  $\underline{r}_1$ .

Figs. 2 and 3 show such a chain reaction apparatus. A neutron radiation, the initial radiation, is generated by the high voltage canal ray tube 1 (shown in greater detail in Fig. 6). This tube generates fast deuterons which strike the target 28 which contains deuterium. The neutron radiation emerging from 28 acts on the matter 3 which fills the spherical transmutation space. The composition of this matter 3 will be discussed further below and is such thet a chain reaction is released by the neutrons. The pumps 120, 121 and 122 pump a liquid, for instance water or mercury through the pipe systems 107, 110, 111 thereby cooling the transmutation area 3 and driving the heated liquid through the boiler 126. The boiler supplies steam to a power plant. The neutrons emerging from the sphere 3 act on a layer 9 which is composed of an element xx that will transmute into a radio-active body (which is suitable for the
 storage of energy).

An essentially different way of introducing the 3 initial radiation into the chain reaction chamber is the 4 arrangement shown in Fig. 4. 401 is the cathode ray 5 tube described in Fig. 1. 402 is a sheet of a heavy 6 element for instance Pb, or U in which penetrating radia-7 8 tion (hard X-rays) is generated with an extremely good efficiency if the electrons have a voltage over one million 9 volts. This efficiency increases very rapidly with the 10 voltage, and is much higher than it could be expected 11 12 from the experience based on ordinary X-ray work. The 13 thickness of the sheet 402 is such as to enable the gener-14 ated penetrating radiation to penetrate through this sheet 15 and act on the transmutation chamber 106 (in Fig. 3). 16 Nevertheless the sheet can be sufficiently thick to utilize 17 more than half of the energy of the cathode rays. The 18 X-rays emerging from sheet 402 penetrate the layer 3 and 19 can liberate neutrons either from the layer 3 or from a 20 substance 407 placed in the interior of the layer 3). For 21 instance, if we have beryllium present in 403 or in 3 22 neutrons will be liberated by X-rays. These neutrons can 23 then maintain a chain reaction as discussed further above and further below. The advantage of using X-rays as an 24 initial radiation is the following: The X-rays penetrate 25 26 through a perfectly closed layer 3 into the interior of 27 the layer and therefore a leak of neutrons from the inter-28 ior can be avoided. This is specially important if we have 29 to deal with a neutron chain in which no multiplicator 30 action is involved. In such cases X-rays may be used with

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advantage as initial radiation especially in view of the
 unexpectedly large efficiency of the X-ray production by
 means of fast electrons acting on heavy elements.

4 We wish to discuss now certain features which arise when the above teachings of this application are 5 applied to uranium. If uranium is exposed to neutrons of 6 7 a few million volts energy or to neutrons slowed down by 8 water to a few volts of energy or less, uranium emits 9 neutrons in two different ways. This can be demonstrated 10 with particular ease if uranium is exposed to slow neutrons 11 and the slow neutron stream hitting the uranium is suddenly 12 stopped. One then finds that uranium emits neutrons after 13 the stoppage of the slow neutron stream for another few, 14 perhaps ten to fifteen, seconds. This delayed neutron 15 emission is weak, i.e. there are less than perhaps thirty 16 neutrons emitted in this way per incident neutron which 17 disappears in reacting with the uranium. No chain reactions 18 could be based on this delayed neutron emission alone. On 19 the other hand, it is easily demonstrated that uranium emits 20 a large number of neutrons while it is exposed to the slow 21 neutron stream and the number of neutrons which are thus 22 instantaneously (within a fraction of a second) emitted 23 from uranium per number of incident neutrons which are 24 captured in the reaction with uranium is larger than one. 25 This fact makes it possible to use uranium as the carrier 26 of a chain reaction. The neutrons which are emitted from 27 uranium during the irradiation with slow neutrons can be 28 distinguished from the incident slow neutrons by virtue 29 of the fact that their velocity is much higher and ranges 30 somewhere between a few ten thousand volts to perhaps about

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1 a million volts or so.

In spite of its small value the delayed neutron emission is not without significance. In view of its existence we have - in order to be more precise - to distinguish between the critical thickness for the total neutron emission (instantaneous and delayed emission together).

7 It is possible to use uranium in the chain reaction 8 layer not only as uranium metal or as uranium oxide but also 9 it is possible to use either of these mixed with a hydrogen 10 containing substance such as for instance water and it is 11 possible to use such a large concentration of the hydrogen 12 containing substance that most of the neutrons are slowed 13 down to a few volts before they react with the uranium in 14 releasing an additional number of neutrons (releasing more 15 than one neutron on the average for every neutron captured by 16 uranium). That the use of hydrogen containing substances in 17 such concentrations should be possible is very surprising 18 and unexpected and is in contradiction with the generally 19 accepted value of the capture cross-section of uranium 20 for neutrons slowed down by water to a few volts velocity 21 or less. The total cross-section of uranium has been 22 assumed on the basis of published measurements to be above 40 x  $10^{-24}$ , i.e. the capture cross-section well above 30. 23 24 On the other hand, the cross-section for that transmutation 25 of uranium which yields on the average neutrons in excess 26 of one but certainly not in excess of 8 per transmutation is 27 known to be less than 3. In these circumstances most of the 28 neutrons which are slowed down to a few volts or less will be 29 captured by uranium without causing a transmutation of the type 30 which yields neutrons. Accordingly one would expect that

less than one neutron will be emitted on the average for 1 every neutron captured by standard uranium. (By "standard 2 uranium" we mean uranium in which the relative abundance 3 of the two isotopes 135 and 138 has the ratio which one 4 finds in uranium that occurs in nature, i.e. about 1:139.) 5 The only possible explanation is that for some reason or 6 other an error has been made by those who previously meas-7 -24 ured the total cross-section or uranium to be about 43 x 10 8 though it is not possible to state through what mistake such 9 10 error occurred.

The mixing of uranium with a hydrogen containing 11 substance to form the chain reaction layer has the advantage 12 13 greatly to reduce the critical thickness and also the amount of uranium required. The uranium or uranium oxide can either 14 15 be mixed with the hydrogen containing substance or the chain 16 reaction layer may be built up from alternating layers of 17 uranium and the hydrogen containing substance. As a 18 hydrogen containing substance water, paraffin wax or calcium hydride appear to be suitable. If the hydrogen containing 19 substance is not mixed with uranium, but alternating layers 20 21 are used, the layers should be as thin as possible. In 22 particular the thickness of the hydrogen containing layer 23 should be as small as possible, and if paraffin, water, or 24 a hydrogen containing substance of about the same hydrogen 25 concentration is used, the thickness of the layer should not 26 exceed about 7 mm.

It is important that the ratio of the number of grams of hydrogen to the number of grams of uranium in the chain reaction layer shall not exceed a certain critical value as otherwise no chain reaction can be maintained for any thickness of the layer which contains the mixture.

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This is due to the fact that hydrogen captures slow neutrons 1 and therefore competes with uranium. The capture cross-2 section of the hydrogen atom is about one-third  $10^{-24}$ 3 and the capture cross-section of uranium for the trans-4 mutations which lead on the average to the liberation of 5 about one additional neutron is about  $2 \times 10^{-24}$  sq. c. 6 7 If these approximate values were exactly correct, no chain 8 reaction would be possible in a layer which contains 9 hydrogen and uranium in a ratio of 6 grams of hydrogen to 10 238 grams of uranium. It is certain that no chain reaction 11 is possible in a layer which contains 20 grams of hydrogen 12 for every 240 grams of uranium. On the other hand, it is 13 desirable in order to keep the critical thickness low 14 not to use too little hydrogen. A reasonable value for 15 the ratio of hydrogen to uranium which gives good results 16 and none too large critical thickness can be found if the 17 expression

18 (7)

XV 1-2-3 ×

19 is plotted and a value of Xm is selected for which the 20 expression becomes maximum. If a mixture is prepared in 21 which the ratio of the number of hydrogen atoms to the 22 number of uranium atoms  $N_n/N_u$  has a value between 23 (8)  $\frac{1}{2} \times m$  and  $\frac{\times m+6}{2}$ 

and if such a mixture is used to build up the chain
reaction layer, satisfactory results will be obtained. As
stated before, alternating layers of the hydrogen containing substance and uranium or uranium oxide can be used
instead of a mixture.

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If instead of standard uranium, uranium is used in 1 which the isotope 235 has been concentrated so that its 2 relative abundance is larger than one part in 139, a larger 3 concentration of hydrogen can be used in the chain reaction 4 layer. The ratio of the hydrogen to the uranium concen-5 tration can be increased by the same factor by which the 6 relative abundance of the uranium isotope 235 has been 7 Such an increase of the relative abundance of increased. 8 the uranium isotope 235 can be achieved by subjecting 9 uranium hexa fluoride to one of the known diffusing pro-10 cesses which lead to separation of molecules of different 11 molecular weight. 12

In such a chain reaction layer which contains such 13 large concentrations of hydrogen as indicated above, the 14 neutron emitted from the uranium in the chain reaction 15 is slowed down after traveling in the chain reaction layer 16 an average distance b from its origin. At the distance 17 b from its origin the neutron will therefore be so slow that 18 its mean free path  $\underline{a}$  for scattering is much smaller than the 19 value b. In paraffin wax this mean free path a would be 20 about 2-1/2 mm, and in the chain reaction layer it will be 21 comewhat larger, i.e. by a factor k which gives the ratio 22 of the concentration of hydrogen in paraffin to the concen-23 tration of hydrogen in the chain reaction layer. This 24 reduction of the mean free path for scattering is due to 25 the large scattering cross-section of hydrogen for room 26 temperature neutrons. In addition, the neutrons which 27 have been slowed down at an average distance b from their 28 origin have now, being slow, a large cross-section for 29 those transmutations of uranium which lead to the liber-30

1 ation of neutrons. The combined effect of the large scattering cross-section of the slow neutrons in the chain reaction 2 layer and the large transmutation cross-section of the slow 3 neutrons for uranium is that a neutron which is emitted by 4 uranium and which becomes slow at the average distance b 5 6 from its point of origin will transmute a uranium nucleus 7 at a point which is at an average distance q from the point 8 at which the neutron became slow, and the distance q is 9 small compared to b, so that the neutron will transmute 10 a uranium nucleus at a distance  $\underline{b}_1$  not very different from 11 b from the point of its origin.

12 In these circumstances the previously given equation 13 no longer holds, and the critical thickness is approximately 14 given in the following way: For the stationary state the 15 neutron density within the spherical chain reaction layer 16 obeys now the equation:

 $(\vec{b}) \frac{d^2(r_0)}{dr^2} + \frac{6(w_2 - w_0)}{w_1 + 2w} (r_1)$ 17 (9) In this equation  $\underline{w}_{o}$  is the probability for a slowed 18 down neutron to cause a transmutation of uranium in which 19 the slow neutron disappears and no fast neutron is emitted; 20  $\underline{w}_1$  is the probability for a slowed down neutron to cause a 21 transmutation of uranium in which the slow neutron disappears 22 and one fast neutron is emitted;  $\underline{w}_2$  is the probability for a 23 slowed down neutron to cause a transmutation of uranium in 24 which the slow neutron disappears and two fast neutrons are 25 emitted. 26

For the special case:  $\underline{w}_2 = 1$ ;  $\underline{w}_1 = 0$ ;  $\underline{w}_0 = 0$ , the above equation gives

29

30 10 
$$(\bar{b})^2 \frac{d^2(rs)}{dr^2} + 3(rs) = 0$$

The critical thickness is given by  $f_{st} = \frac{1}{3} \frac{1}{6} \left( \frac{w_2 - w_0}{3(w_1 - 2w_2)} \right)^{1/2}$ (1)2 in the general case and by 3

(12)  $l_{0} = \frac{\pi}{2} \vec{b} V = \frac{\pi}{2}$ 

5 in the special case.

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6 Obviously, the above diffusion equation presupposes for its validity a small value of  $\underline{w}_2$ , but even 7 for large values of  $\underline{W}_2$  it gives at least the order of 8 9 magnitude for the critical thickness.

10 The critical thickness will in practice always 11 be determined empirically for instance in the following 12 way: a neutron source is surrounded by the chain reaction 13 layer of an approximately correct thickness which is safely 14 below the critical thickness. The radiations emitted from 15 the chain reaction layer, while exposed to this neutron 16 source are observed by means of an ionization chamber. 17 Then the thickness of the chain reaction layer is brought 18 closer to the critical thickness by gradually increasing 19 either the quantity of uranium or the quantity of hydrogen 20 containing substances mixed with the uranium. The amount 21 of ionizing radiation which is emitted is again observed 22 and the thickness of the chain reaction layer is again 23 brought closer to the critical thickness in the same way 24 as before. In this way, by observing the increase of the 25 emitted radiation as a function of the increasing effect-26 ive thickness of the chain reaction layer the critical 27 thickness can be extrapolated from the observed curve by 28 plotting the intensity of the emitted neutron radiation 29 against the effective thickness of the chain reaction 30 layer. Instead of an ionization chamber which registers

the neutron intensity by means of recoil ions in the gas of the chamber, induced activity caused by the neutrons can be used as a measure of the radiation intensity.

4

## Variation of Critical Thickness

If slow neutrons are used the critical thickness can 5 be increased by having a slow neutron absorber within the 6 hollow sphere in the center of the spherical arrangement. 7 If the inner radius of the spherical shell of the chain 8 reaction layer is much larger than the critical thickness 9 (to be accurate we have said the standard critical thick-10 11 ness given by the above formulas), and if all slow neutrons 12 are absorbed. for instance by a cadmium layer covering the 13 inner surface of the spherical chain reaction layer, the critical thickness of the arrangement is increased. 14 Bv 15 removing such absorbing matter from the inside of the chain 16 reaction layer, the critical thickness may be reduced below 17 the actual thickness, and thus an explosion may be brought 18 The explosion will be all the more violent the about. 19 more quickly the absorbing substance is removed. A similar 20 increase in the critical thickness of a spherically sym-21 metrical chain reaction layer can be brought about by 22 removing a section of the layer and thereby producing an 23 opening through which the neutrons can escape. For instance 24 a conical section corresponding to a few percent of the 25 spherical chain reaction layer can be so arranged as to be 26 easily moved out of its place and replaced, and thereby the 27 critical thickness may be reduced or increased.

Explosions may be brought about by a variation of the critical thickness for the purpose of creating a large amount of radio active elements. In such a case it is

desirable to have mild explosions. Mild explosions can 1 be brought about by slowly changing the space distri-2 3 bution of matter and also by arranging layers so that when the explosion sets in there shall be no rapid 4 5 removal of substances which capture neutrons and thereby 6 reduce the critical thickness. For instance, if the 7 chain reaction layer is based on fast neutrons and we have 8 a substance in the hollow sphere in the center of the chain 9 reaction layer which will slow down neutrons and absorb 10 the slow neutrons, for instance a solution of a boron 11 salt in water, the arrangement may be such that there shall 12 be no easy outflow for the water from the inside of the 13 spherical chain reaction layer towards the outside. On 14 the contrary, if a strong destructive explosion is wanted, 15 it can be brought about by providing for such outflow. 16 It is advisable to have explosions which for the purpose 17 of reducing radio active elements are so arranged that 18 the explosions shall take place in the middle of a large 19 water tank or below water. After the explosion the 20 scattered radio active material can then be collected 21 from the water.

### Regulation

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23 As we have seen, the ratio of neutron input to 24 neutron output becomes infinite for the critical thickness 25 of the arrangement. The neutron input is in practice limited by the accuracy of the arrangement, since the 26 27 thickness of the chain reaction layer must be extremely 28 close to the critical thickness, and yet must remain below 29 it in order to avoid an explosion. Fortunately, it is 30 possible to overcome this difficulty by reason of the

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## 1 following fact:

In reality we have to deal not with one critical 2 thickness only, but with two different critical thicknesses 3 which we shall call the instantaneous critical thickness 4 and the delayed critical thickness. They arise by virtue 5 of the fact that, while the bulk of neutrons is emitted 6 7 instantaneously when uranium is transmuted by neutrons, there is also a delayed emission of neutrons, the delay 8 9 being of the order of magnitude of a few seconds. If the thickness of the chain reaction layer is larger than the 10 delayed critical thickness, but smaller than the instan-11 taneous critical thickness, the neutron output increases 12 13 to infinity, but does not increase too rapidly. This 14 makes it possible that by moving an object which forms part 15 of the arrangement and which has an influence on the critical thicknesses (for instance, by having a slow neutron absorber 16 in the interior of the hollow sphere of the spherical chain 17 18 reaction layer, and by partially withdrawing it from there. 19 we can reduce the critical thickness, and in a similar way 20 we can also increase it by the opposite movement), we can 21 vary the critical thickness in time. We shall call objects which are used in this way regulator objects, and accord-22 23 ing to our invention the neutron output can be kept very 24 high without risking an explosion by moving the regulator 25 object in such a way that part of the time the critical 26 thickness for delayed emission should be below, and part 27 of the time it should be above, the real thickness of the 28 spherical layer. It is only necessary for safe functioning 29 to have an instrument which is sensitive to the emitted radi-30 ation or the temperature of some part of the chain reaction

layer, and this instrument can control the position of 1 the regulator object. Obviously, in order to have stable 2 functioning, the regulator object will have to be moved in 3 direction of an increase of the critical thickness with 4 increasing neutron radiation, and it has to be moved in the 5 opposite way with decreasing neutron radiation. While the 6 7 thickness of the chain reaction layer will still have to be 8 accurately chosen, since it has to be within narrow limits, i.e. between the critical thickness for instantaneous 9 neutron emission and the critical thickness for delayed 10 11 neutron emission, the latter being only slightly larger 12 than the former. Yet the above described regulation makes 13 it possible to get a very much higher neutron output without 14 reaching an explosion.

15 The critical thickness for delayed emission could 16 also be conveniently called the total critical thickness 17 because it corresponds to the total neutron emission, both 18 instantaneous and delayed. It can be easily determined 19 empirically by varying the thickness of the chain reaction 20 layer and observing for each thickness the emitted neutron 21 radiation as a function of time. Below the critical thick-22 ness for delayed emission the neutron radiation is a function 23 of time which resembles a growth curve in the field of radio 24 activity, i.e. it approaches an upper limit practically 25 reaching saturation after some time. Above the critical 26 thickness for delayed emission, but below the critical thick-27 ness for instantaneous emission, the observed neutron emission increases more and more rapidly with time, and the 29 arrangement has quickly to be changed in order to avoid 30 overheating. The value of the critical thickness for

1	delayed emission is reached when one type of curve goes
2	over into the other, and at the critical thickness itself
3	the neutron intensity as a function of time is a straight
4	line.
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We wish now to insert a description of Figs. 5, 6 and 7 to which we have referred in the preceding part of the specification.

Fig. 5 shows an example of a suitable arrangement. 4 11 is an electrical discharge tube ejecting a beam 12 of 5 fast diplogen ions.\* The ions fall on a substance 13 6 consisting of for instance gaseous diplogen or a diplogen 7 compound or lithium, causing transmutation, i.e. a nuclear 8 reaction of the diplogen ion with an atom of the target. 9 The substance 13 is surrounded by a layer 14 containing 10 the element which we wish to transmute into a radio active 11 element. In order to have a good efficiency, the thick-12 ness of the layer 14 has to be sufficiently large, compared 13 14 with the mean free path of the neutron for this transmutation. 15

Fig. 6 shows the electrical discharge tube referred
to in Fig. 5. It is a high voltage positive ray tube.
There is an auxiliary positive ray tube on top of the high
voltage tube. 11 is the anode, 15 the cathode of this
auxiliary tube. Diplogen is admitted through the tube 13
and pumped away through

Fig. 7 shows an arrangement suitable for the 22 23 production of hard X-rays. 1 is the primary of a transformer, the secondary 2 of which is connected to the points 24 3 and 4. 3 is connected to the cathode 8 of the rectifier 25 tube 5 and to the anode 7 of the rectifier tube 6. Point 4 26 27 is connected to the cathode 9 of the rectifier tube 10 and to the anode 11 of the rectifier tube 12. The cathodes 13 28 and 14 are connected to each other and to the earth. 29 The 30 anodes 15 and 16 are connected to point 17, and this point

also called diplons or deutons

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is connected to the pole 18 of the impulse generator 20, the
 pole 19 of which is connected to earth. The impulse gener ator 20 is built of condensers 21, resistances 22 and spark
 gaps 23.

This impulse generator is adapted to produce inter-mittent voltage up to 10 million volts, transmitted to the discharge tube 24 through the spark gap 25. 26 is the cathode of the discharge tube, the anode 27 of which is connected to the earth. The fast electrons emerge through the metal window 27 (which is the anode as well) and hit a body 28. This body is used as an anticathode and yields hard X-rays with very good efficiency if it is built of Bi, Pb or some other heavy element.

The invention concerns a process and apparatus for the transmutation of elements and makes it possible to bring about such a transmutation on a large scale. The production of power by means of the heat generated in the process and the production of radio active elements on a large scale are thus made possible by the invention. The invention teaches that it is possible to produce a nuclear chain reaction and to maintain the stationary conditions in such chain reactions. It also teaches that it is possible to have explosive bodies in which an explosion is brought about at will by a sudden change in the distribution of matter.

According to the invention such a chain reaction may be maintained for instance by maintaining an initial radiation of neutrons by means of one of the known methods for the production of neutron radiations (for instance by bombarding a lithium target with deuterons or by exposing beryllium to X-rays or gamma-rays) and letting these neuof trons fall on a body/suitably chosen substance, shape and dimensions and thereby obtaining an increase in the number and energy of the neutrons which increase is brought about by their interaction with the said body. Thus if the proper conditions are observed the interaction of neutrons with matter can lead to the liberation of further neutrons. These newly liberated neutrons liberate again in their turn further neutrons so that we have a chain reaction in which a large number of neutrons are liberated, the total number being limited only by the dimensions of the geometrical arrangement.



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Such change is not only possible in the presence of a meta stable element. A meta stable element is an element the mass of which (packing fraction) is sufficiently high to allow its disintegration into its parts and a liberation of energy. Elements like uranium and thorium are examples of such elements. It is possible to measure the packing fraction of the elements by means of the mass spectograph and thereby determine whether an element is meta stable or not in the sense in which the word is used in this application. If, for instance, the values which were generally accepted in 1934 for helium and beryllium had been correct (and we know today that they were false, at least in their relation to each other), we have been correct in concluding from these values which were accepted in 1934 that beryllium is a meta stable element and can be disintegrated into parts with a liberation of energy, one of the parts set free in its disintegration being a neutron. If such an element which is meta stable but the disintegration of which is inhibited is exposed to neutrons, we may have a reaction in which a neutron disappears and more than one neutron is emitted. The additional neutrons would together with the number of the original neutrons continue to interact with the meta stable element thereby forming the links of a chain reaction.

In order to be able to utilize a nuclear reaction in which an excess number of neutrons is liberated by neutrons for the maintenance of a chain reaction, it is not sufficient to measure the cross-section and other constants of the reaction, but it is also necessary to be aware of the laws which govern the neutron output of such reactions in function of the geometrical conditions. Once the general laws, the

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As a carrier of such a chain reaction it is necessary to have an element the mass of which is sufficiently high to allow its disintegration into its parts with liberation of energy, the disintegration being ordinarily inhibited (meta stable state), the inhibition being lifted in a reaction with a neutron. An example of such a meta stable element is uranium.

In order to be able to utilize a nuclear reaction in which an excess number of neutrons is liberated by neutrons for the maintenance of a chain reaction, it is not sufficient to measure the cross-section and other constants of the reaction, but it/also necessary to be aware of the laws which govern the neutron output of such reactions in function of the geometrical conditions. Once the general laws, the type of behavior is known, the exact dimensions can be easily determined in each particular case by actually measuring the neutron output. Such experimental adjustments can, however, only be made if the general type of behavior is known.

In the following we shall by way of example demonstrate certain general features of such a neutron chain reaction in a special case. In this special case the carrier of the chain reaction forms a spherical layer as illustrated in Fig. 1. In Fig. 1, 1 is a spherical layer which contains a carrier of the chain reaction, 2 is a neutron source which has fixed constant output of neutrons,  $\underline{r}_1$  is the inner radius of the spherical layer. In order to be able to treat the problem as a diffusion problem, we choose  $\underline{r}_1$  very much larger than the mean free path <u>a</u> of the neutrons produced in the chain reaction in the substance of

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the chain reaction layer. The neutrons liberated in the chain reaction layer will have to make a number of collisions with the nuclei which compose the chain reaction layer, and we designate the number of collisions necessary for causing transmutations in the carrier of the chain reaction layer which lead on the average to the liberation of one additional neutron with  $\underline{f}$ . We further assume that  $\underline{f}$  is a large number so that we may have conditions in which the well known equations of diffusion can be applied. With these assumptions the density  $\underline{s}$  of the neutron will with good approximation be given within the chain reaction layer as a function of the radius  $\underline{r}$  by the following equation: (1)  $D \frac{d^2(\underline{r},\underline{s})}{d\tau^2} \neq A(\tau s) = 0$ 

D is a diffusion constant which is given by

D= aw

<u>w</u> is the mean velocity of the neutrons; <u>a</u> the mean free path for scattering collisions within the chain reaction layer; and <u>f</u> a number which says how many scattering collisions a neutron has to make in the chain reaction layer in order to produce on the average an additional neutron.

<u>A</u> stands for the number of neutrons produced per c.c. of chain reaction layer in a second and its value is given by  $A = \frac{\omega}{\alpha f} \quad \text{so that } \sqrt{\frac{D}{A}} = \frac{\alpha \sqrt{f}}{\sqrt{2}}$ 

For a given value of  $\underline{r}_1$  there is a certain value of  $\underline{r}_2$ and accordingly a certain value of  $\underline{r}_2 - \underline{r}_1$  for which the number of neutrons per second diffusing out of the spherical layer into space becomes infinite for a finite neutron production of the source. This value of  $(\underline{r}_2 - \underline{r}_1)$  we may call the critical thickness of the chain reaction layer. If the



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thickness of the chain reaction layer is smaller than but very close to the critical thickness, the neutron output is very much larger than the neutron input and we may have 1000 or more times as many neutrons emerging from the chain reaction layer as the neutron input of the neutron source 2. The value of the critical thickness is a function of ry and of the boundary conditions for  $\underline{r} = \underline{r}_1$  and  $\underline{r} = \underline{r}_2$ . If the outer surface  $(\underline{r} = \underline{r}_2)$  of the spherical layer were to stand free in space, the density s would be 0 for the outer surface, and if there is no absorption of neutrons in the hollow sphere containing the neutron source, i.e. if the number of neutrons produced by the neutron source is equal to the number of neutrons diffusing outwards from the sphere  $\underline{r} = \underline{r}_1$ , we obtain very large values of  $\underline{r}_1$   $(\underline{r}_1 >> \sqrt{\frac{3}{4}})$ ) for the value of the critical thickness (2)  $L_{st} = \frac{\pi}{2} \left| \frac{D}{A} \right| = \frac{\pi}{2} \frac{Q}{\sqrt{2}} \left| \frac{D}{3} \right|$ (2)

The critical thickness for these conditions we shall call further below the standard critical thickness of the substance which composes the chain reaction layer.

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If the outer surface is covered by some material, for instance if the transmutation layer is covered by lead or immersed into water, the critical thickness is less than the standard critical thickness. This is due to the back scattering by lead or water, and, in the case of water, the fact that the neutrons are slowed down by water and their mean free path is thereby reduced plays an important role.

If the neutrons are allowed to escape out of the hollow sphere containing the neutron source in the interior of the spherical layer or if they are absorbed within this hollow sphere, the critical thickness is increased. If all the neutrons were captured in the hollow sphere within  $\underline{r} = \underline{r}_1$ , the critical thickness would become independent of the value of  $\underline{r}_1$  and have a value exactly double the normal critical thickness provided that the neutron density  $\underline{s}$  remains 0 at the outer surface ( $\underline{r} = \underline{r}_2$ ).

Obviously the above given diffusion equation holds only for stationary solutions, that is for solutions where the neutron density  $\underline{s}$  is a function of  $\underline{r}$  only and does not vary with time. Not for all boundary conditions will such stationary solutions exist. If we gradually increase that thickness of the **Aritical** layer we reach for any given boundary conditions a thickness at which the neutron outflow becomes infinite for finite neutron production of the  $\underbrace{flow}_{le}$  restrict thickness is  $\cancel{s}$  critical thickness of the arrangement. It can be calculated for every case in the following way from given boundary conditions.

The solution of the above given diffusion equation has the form of (3)  $sr = C_1 sin(\overline{A}^T) + C_2 cos(\overline{A}^T)$ 

For the boundary condition  $\underline{s} = 0$  for  $\underline{r} = \underline{r}_2$  the solution takes the form of (4)  $Sr = C Sin [r_2 - r] [A]$ 

s = C sin (n-r) 10/4-7

If there is no absorption of neutrons in the hollow sphere ground the neutron source and if the neutron source produces a fixed number N of neutrons per second, the number of neutrons diffusing from the sphere  $\underline{r} = \underline{r}_1$ into the chain reaction layer must be equal to N. On the other hand, the number of neutrons thus diffusing through

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or (5) any sphere is given by  $4\pi r^2 D \frac{ds}{dr}$ 

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and it is therefore also  $N = 4\pi \tau^2 D \frac{ds}{dr} X + \frac{ds$ 

For a value of  $\underline{\mathbf{r}} = \underline{\mathbf{r}}_{\mathbf{n}}$  for which  $\underbrace{df}_{dT}$  becomes 0, the ratio of the number of neutrons diffusing through the outer surface  $\underline{\mathbf{r}} = \underline{\mathbf{r}}_2$  to N becomes infinite. The corresponding which value of  $(\underline{\mathbf{r}}_{\mathbf{n}} - \underline{\mathbf{r}}_1)$  is therefore the critical thickness and the standard for the values of  $\underline{\mathbf{r}}_1$ . For very small values of  $\underline{\mathbf{r}}_1$  ( $\mathcal{T}_1 \ll \left\lfloor \frac{D}{A} \right\rfloor$ ) it obviously becomes twice the standard critical thickness  $\mathcal{L} \cdot \mathcal{C} \cdot \mathcal{T} / \frac{D}{A}$ 

To take another example, if the boundary conditions are  $\underline{s} = 0$  both for  $\underline{r}_1$  and  $\underline{r}_2$ , then a glance at equation (5) shows that  $(\underline{r}_2 - \underline{r}_1) = \overline{n} / \frac{D}{A}$ , i.e. twice the critical thickness, quite independent of the value of  $\underline{r}_1$ .

any sphere is given by  $4\pi r^2 D \frac{ds}{dr}$ 

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of the number of neutrons diffusing through the outer surface  $\underline{r} = \underline{r}_2$  to N becomes infinite. The corresponding value of  $(\underline{\mathbf{r}}_{\mathbf{d}} - \underline{\mathbf{r}}_{\mathbf{l}})$  is therefore the critical thickness and (5)where the critical thickness are the critical thickness and (5)where the critical the critical thickness are the critical the small values of  $\underline{r_i}$  ( $T_i \leftarrow \left(\frac{D}{A}\right)$  it obviously becomes twice the standard critical thickness  $\dot{L} \cdot e \cdot \mathcal{T} = \left(\frac{D}{A}\right)$ 

To take another example, if the boundary conditions are  $\underline{s} = 0$  both for  $\underline{r}_1$  and  $\underline{r}_2$ , then a glance at equation (5) shows that  $\underline{r}_2 - \underline{r}_1 = \pi / \frac{D}{A}$ , i.e. twice the critical thickness, quite independent of the value of r1.

Figs. 2 and 3 show such a chain reaction apparatus. A neutron radiation, the initial radiation, is generated by the high voltage canal ray tube 1 (shown in greater detail in Fig. ). This tube generates fast deuterons which strike the target 28 which contains deuterium. The neutron radiation emerging from 28 acts on the matter 3 which fills the spherical transmutation space. The composition of this matter 3 will be discussed further below and is such that a chain reaction is released by the neutrons. The pumps 120, 121 and 122 pump a liquid, for instance water or mercury, through the pipe systems 107, 110, 111 thereby cooling the transmutation area 3 and driving the heated liquid through the boiler 126. The boiler supplies steam to a power plant. The neutrons emerging from the sphere 3 act on a layer 9 which is composed of an element xx that will trans-

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mute into a radio-active body (which is suitable for the storage of energy).

An essentially different way of introducing the initial radiation into the chain reaction chamber is the arrangement shown in Fig. 4. 401 is the cathode ray tube described in Fig. 1. 402 is a sheet of a heavy element for instance Pb, or U in which penetrating radiation (hard X-rays) is generated with an extremely good efficiency if the electrons have a voltage over one million volts. This efficiency increases very rapidly with the voltage, and is much higher than it could be expected from the experience based on ordinary X-ray work. The thickness of the sheet 402 is such as to enable the generated penetrating radiation to penetrate through this sheet and act on the transmutation chamber 106 (in Fig. 3). Nevertheless the sheet can be sufficiently thick to utilize more than half of the energy of the cathode rays. The X-rays emerging from sheet 402 penetrate the layer 3 and can liberate neutrons either from the layer 3 or from a substance 407 placed in the interior of the layer 3). For instance, if we have beryllium present in 403 or in 3 neutrons will be liberated by X-rays. These neutrons can then maintain a chain reaction as discussed further above and further below. The advantage of using X-rays as an initial radiation is the following: The X-rays penetrate through a perfectly closed layer 3 into the interior of the layer and therefore a leak of neutrons from the interior can be avoided. This is specially important if we have to deal with a neutron chain in which no multiplicator action is involved. In such cases X-rays may be used with

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advantage as initial radiation especially in view of the unexpectedly large efficiency of the X-ray production by means of fast electrons acting on heavy elements.

We wish to discuss now certain features which arise when the above teachings of this application are applied to uranium. If uranium is exposed to neutrons of a few million volts energy or to neutrons slowed down by water to a few volts of energy or less, uranium emits neutrons in two different ways. This can be demonstrated with particular ease if uranium is exposed to slow neutrons and the slow neutron stream hitting the uranium is suddenly One then finds that uranium emits neutrons after stopped. the stoppage of the slow neutron stream for another few, perhaps ten to fifteen, seconds. This delayed neutron emission is weak, i.e. there are less than perhaps thirty neutrons emitted in this way per incident neutron which disappears in reacting with the uranium. No chain reactions could be based on this delayed neutron emission alone. On the other hand, it is easily demonstrated that uranium emits a large number of neutrons while it is exposed to the slow neutron stream and the number of neutrons which are thus instantaneously (within a fraction of a second) emitted from uranium per number of incident neutrons which are captured in the reaction with uranium is larger than one. This fact makes it possible to use uranium as the carrier of a chain reaction. The neutrons which are emitted from uranium during the irradiation with slow neutrons can be distinguished from the incident slow neutrons by virtue of the fact that their velocity is much higher and ranges somewhere between a few ten thousand volts to perhaps about

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a million volts or so.

In spite of this small value the delayed neutron emission is not without significance. In view of its acceptance we have to be precise to distinguish between the critical thickness for the total neutron emission (instantaneous and delayed emission together).

#### I CLAIM:

1. Production of radio active elements comprising the step of maintaining a neutron radiation in a layer containing uranium, the thickness of the said layer being slightly below the critical thickness.

2. Production of radio active elements comprising the step of maintaining a neutron radiation in a layer containing uranium, the thickness of the said layer being slightly below the critical thickness, and the step of separating at intervals from the layer the radio active elements produced in the layer.

3. Production of radio active elements comprising the step of maintaining a neutron radiation in a layer containing uranium, the thickness of the said layer being slightly below the critical thickness, and exposing another element which can be made radio active by means of slow neutrons to the radiation of the chain reaction layer.

4. Production of radio active elements according to claims 1 to 3 comprising the step of maintaining a neutron radiation in a layer containing uranium or uranium oxide, the thickness of the layer being chosen so as to conform with equation (1) and given by equation (5) according to the prescription (6) respectively using values for the mean free path corresponding to a cross-section of uranium of about 6 x  $10^{-24}$  and oxygen of about 3 x  $10^{-24}$ and a value of <u>f</u> of about 60.

5. Production of radio active elements according to claim 4, the layer being composed of metallic uranium and having a thickness of about 30 to 70 centimeters for an inner radius  $\underline{r}_1$  about 10 centimeters.





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6. Production of radio active elements according to claim 5, the spherical chain reaction layer of metallic uranium being surrounded with a spherical layer of metallic bismuth the thickness of which bismuth layer exceeds 50 centimeters and the thickness of the uranium layer being below the values given in claim 5, the thickness of uranium being reduced to about half if very large amounts of bismuth are used.

7. Production of radio active elements according to claims 1 to 3 in which a hydrogen containing substance, such as for instance paraffin wax or water, is mixed with uranium, the number of gram atoms of hydrogen having the ratio to the number of gram atoms of uranium given by equation (8)

8. Production of radio active elements according to claim 7, the chain reaction layer being surrounded with a spherical layer of bismuth metal exceeding in thickness 50 centimeters.

9. Production of radio active elements according to claim 7, the spherical chain reaction layer being immersed in water or another similar hydrogen containing substance, for instance paraffin wax.

10. Production of radio active elements according to claim 1 or claim 7 in which the uranium used has a different relative abundance of the isotopes 235 and 238 than standard uranium, the relative abundance of uranium 235 being increased by a factor of 3 or more.

11. Production of radio active elements comprising the step of increasing the critical thickness of an arrangement comprising a layer of uranium, the said increase being effected by changing the distribution of matter within or without the chain reaction layer and the increase carried to the point where the critical thickness of the arrangement is exceeded so that a chain reaction leading to an explosion takes place.

12. Production of radio active elements according to claim 11 in which the process described in claim 11 is carried out in a place surrounded by water or below water and the radio active elements produced through the explosion and scattered in the water are collected.

13. Production of power comprising the step of maintaining a neutron radiation in a layer containing uranium, the thickness of the said layer being slightly below the critical thickness.

14. Production of power according to claim 13 comprising the step of maintaining a neutron radiation in a layer containing uranium or uranium oxide, the thickness of the layer being chosen so as to conform with equation (1) and given by equation (5) according to the prescription (6) respectively using values for the mean free path corresponding to a cross-section of uranium of about 6 x  $10^{-24}$ and oxygen of about 3 x  $10^{-24}$  and a value of <u>f</u> of about 60.

15. Production of power according to claim 14, the layer being composed of metallic uranium and having a thickness of about 30 to 70 centimeters for an inner radius  $\underline{r}_1$  about 10 centimeters.

16. Production of power according to claim 15, the spherical chain reaction layer of metallic uranium being surrounded with a spherical layer of metallic bismuth the thickness of which bismuth layer exceeds 50 centimeters and the thickness of the uranium layer being below the values given in claim 5, the thickness of uranium being reduced to about half if very large amounts of bismuth are used.

17. Production of power according to claim 13 in which a hydrogen containing substance, such as for instance paraffin wax or water, is mixed with uranium, the number of gram atoms of hydrogen having the ratio to the number of gram atoms of uranium given by equation (8).

18. Production of power according to claim 17, the chain reaction layer being surrounded with a spherical layer of bismuth metal exceeding in thickness 50 centimeters.

19. Production of power according to claim 17, the spherical chain reaction layer being immersed in water or another similar hydrogen containing substance, for instance paraffin wax.

20. Production of power according to claim 12 or claim 17 in which the uranium used has a different relative abundance of the isotopes 235 and 238 than standard uranium, the relative abundance of uranium 235 being increased by a factor of 3 or more.

21. Production of power comprising the step of increasing the critical thickness of an arrangement comprising a layer of uranium, the said increase being effected by changing the distribution of matter within or without the chain reaction layer and the increase carried to the point where the critical thickness of the arrangement is exceeded so that a chain reaction leading to an explosion takes place.

#### I CLAIM:

1. Production of power comprising the step of maintaining a neutron radiation in a layer containing uranium, the thickness of the said layer being slightly below the critical thickness.

2. Production of radio active elements comprising the step of maintaining a neutron radiation in a layer containing uranium, the thickness of the said layer being slightly below the critical thickness, and the step of separating at intervals from the layer the radio active elements produced in the layer.

3. Production of radio active elements comprising the step of maintaining a neutron radiation in a layer containing uranium, the thickness of the said layer being slightly below the critical thickness, and exposing another element which can be made radio active by means of slow neutrons to the radiation of the chain reaction layer.

4. Production of power according to claims 1 to 3 comprising the step of maintaining a neutron radiation in a layer containing uranium or uranium oxide, the thickness of the layer being chosen so as to conform with equations (1) and given by equation (5) according to the prescription (6) respectively using values for the mean free path corresponding to a cross-section of uranium of about 6 x  $10^{-24}$  and oxygen of about 3 x  $10^{-24}$  and a value of <u>f</u> of about 60.

5. Production of power according to claim 4, the layer being composed of metallic uranium and having a thickness of about 30 to 70 centimeters for an inner radius  $\underline{r}_1$  about 10 centimeters.



Mr. Leo Syilard To Pennie Davis Marin & Edmonds 165 Broadway n.y.C.

6. Production of power according to claim 5, the spherical chain reaction layer of metallic uranium being surrounded with a spherical layer of metallic bismuth the thickness of which bismuth layer exceeds 50 centimeters and the thickness of the uranium layer being below the values given in claim 5, the thickness of uranium being reduced to about half if very large amounts of bismuth are used.

7. Production of power according to claims 1 to 3 in which a hydrogen containing substance such as for instance paraffin wax or water is mixed with uranium, the number of gram atoms of hydrogen having the ratio to the number of gram atoms of uranium given by equation (8).

8. Production of power according to claim 7, the chain reaction layer being surrounded with a spherical layer of bismuth metal exceeding in thickness 50 centimeters.

9. Production of power according to claim 7, the spherical chain reaction layer being immersed in water or another similar hydrogen containing substance for instance paraffin wax.

10. Production of power according to claim 1 or claim 7 in which the uranium used has a different relative abundance of the isotopes 235 and 238 than standard uranium, the relative abundance of uranium 235 being increased by a factor of 3 or more.

11. Production of power comprising the step of increasing the critical thickness of an arrangement comprising a layer of uranium, the said increase being effected by changing the distribution of matter within or without the chain reaction layer and the increase carried to the point where the critical thickness of the arrangement is exceeded so that a chain reaction leading to an explosion takes place.

12. Production of power according to claim 11 in which the process described in claim 11 is carried out in a place surrounded by water or below water and the radio active elements produced through the explosion and scattered in the water are collected. COPY

#### DEPARTMENT OF COMMERCE

UNITED STATES PATENT OFFICE

WASHINGTON

Please find below a communication from the EXAMINER in charge of this application.

CONWAY P. COE

Applicant: Leo Szilard

Ser. No. 263,017 Filed Mar. 20, 1939 For Apparatus for Nuclear Transmutation.

Commissioner of Patents

Pennie, Davis, Marvin & Edmonds, 165 Broadway, New York, N.Y.

AMENDMENT DUE FEB 26 1940

References:

Bemis	1,588,568	June 15,	1926	60-1
Swartwout	1,641,830	Sept. 6,	1927	60-1
Szilard	2,161,985	June 13,	1939	204-31.02

Division is required between the following groups: Group I - claims 1 to 12, classifiable in 204-31, drawn to a process for the production of radio active elements, and

Group II - claims 13 to 21, drawn to a process for the production of power classifiable in 60-1.

A process for the production of power is obviously separate and independent of one for the production of radio active elements. The two groups are separately classifiable as indicated by the patents above cited and the searches are widely divergent.

The art is cited to illustrate the line of division. Action on the merits is deferred until the question of division is determined.

All the claims are rejected in view of the requirement of division.

EXAMINER.

## DEPARTMENT OF COMMERCE

UNITED STATES PATENT OFFICE

WASHINGTON

OCT 16 1942

MAILED

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Serial No. 263,017

Filed March 20, 1939

For

Apparatus for Nuclear Transmutation

Applicant Leo Szilard

Assignee

NOTICE:- To the applicant above named, his heirs, and any and all his assignees, attorneys and agents:

Under the provisions of the Act of October 6, 1917 (Public No. 80), as amended July 1, 1940 (Public No. 700), as amended August 21, 1941 (Public No. 239), you are hereby notified that your application as above identified has been found to contain subject matter the disclosure of which might be detrimental to the public safety or defense, and you are hereby ordered to in nowise publish or disclose the invention or any hitherto unpublished details of the disclosure of said application, but to keep the same secret (except by written permission first obtained of the Commissioner of Patents), under the penalties of the amended Act. This application must be prosecuted under the Rules of Practice until a notice is received from the office that all the claims then in the case are allowable. Such notice closes the prosecution of the case. Furthermore, if previously allowed and now withdrawn from issue the prosecution of the case is likewise closed. When the application is in condition for allowance it will be withheld from issue during such period or periods as the national interest requires.

This order should not be construed in any way to mean that the Government has adopted or contemplates adoption of the alleged invention disclosed in this application, nor is it any indication of the value of such invention. In order to make the details of your invention available for inspection by the various national defense agencies for defense purposes and at the same time to preserve your rights under the Act, it is suggested that you promptly tender this invention to the Government of the United States for its use. Such tender may be effected by a communication directed to the Secretary of War or to the Secretary of the Navy and should be accompanied by a power to inspect the application and a copy of the application, including drawings.

DATED 0CT 14 1942

Commissioner.

pb





# 2161985

# INFOR UNIVERID STRANKS OF MURICA

#### TO ALL TO WHOM THESE PRESENTS SHALL COME:

Rhereas

LEO SZILARD,

of

New York,

N. Y.,

PRESENTED TO THE Commissioner of Patents a petition praying for the grant of Letters Patent for an alleged new and useful improvement in

PROCESSES OF PRODUCING RADIO-ACTIVE ELEMENTS,

A DESCRIPTION OF WHICH INVENTION IS CONTAINED IN THE SPECIFICATION OF WHICH A COPY IS HEREUNTO ANNEXED AND MADE A PART HEREOF, AND COMPLIED WITH THE VARIOUS REQUIREMENTS OF LAW IN SUCH CASES MADE AND PROVIDED, AND

REFELLAS UPON DUE EXAMINATION MADE THE SAID CLAIMANT IS ADJUDGED TO BE JUSTLY ENTITLED TO A PATENT UNDER THE LAW.

Now therefore these Letters Patent are to grant unto the said

Leo Szilard, his heirs

Attest:

aminer.

OR ASSIGNS

FOR THE TERM OF SEVENTEEN YEARS FROM THE DATE OF THIS GRANT

THE EXCLUSIVE RIGHT TO MAKE, USE AND VEND THE SAID INVENTION THROUGHOUT THE UNITED STATES AND THE TERRITORIES THEREOF.

Intestimony whereof Thave hereunto set my hand and caused the seal of the Patent Office to be affixed at the City of Washington this thirteenth day of June, in the year of our Lord one thousand nine hundred and thirty-nine, and of the Independence of the United States of America the one hundred and sixty-third.

E 13-1 V - Sigulard 3161 - 985 

June 13, 1939.

L SZILARD

2,161,985



Freentor:

### June 13, 1939.

2,161,985

PROCESS OF PRODUCING RADIC-ACTIVE ELEMENTS

Filed March 11, 1935

3 Sheets-Sheet 2



Freenton:

### June 13, 1939. L SZILARD

2,161,985

PROCESS OF PRODUCING RADIC-ACTIVE ELEMENTS Filed March 11, 1935 3 Sheets-Sheet 3

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Inventor:

Les holand

Patented June 13, 1939

2,161,985

### UNITED STATES PATENT OFFICE

#### 2,161,985

PROCESS OF PRODUCING RADIO-ACTIVE ELEMENTS

Leo Szilard, New York, N. Y.

Application March 11, 1935, Serial No. 10,500 In Great Britain March 12, 1934

9 Claims. (Cl. 204-31)

This invention concerns methods and apparatus for the generation of radio-active bodies.

- According to one feature of my invention, radio-active elements may be produced from nattor compounds of natural elements with neutrons produced in various ways, more particularly, by subjecting the natural elements to neutrons emanating from a target containing lithium, which
- 10 target is subjected to a bombardment with fast deuterons. Another feature of the invention is directed to the production of radio-active elements from natural elements by exposing the natural elements to an irradiation with neutrons
- 45 which are liberated from certain elements under the action of X-rays. Another feature of the invention is directed to chemically concentrating radio-active elements produced from natural elements if the radio-active element is isotopic
  20 with the natural element from which it is pro-
- duced. Other features of the invention will appear in the following detailed description referring to the drawings, and will be more particularly pointed

25 out in the claims.

In the drawings,

Figure 1 represents a sectional elevation of an apparatus for carrying out the invention,

- Figure 2 shows a more constructional lay-out 30 of the apparatus of Figure 1,
  - Figure 3 shows the circuit arrangements for further modified apparatus and,
    - Figure 4 is a sectional view of apparatus intended to co-operate with that shown in Figure 3.
- Referring first to Figure 1 of the drawings, 11
   is an electrical discharge tube adapted to project a beam 12 of fast deuterons. The tube 11
   is filled with deuterium and an anode A and cathode B are provided for connection to a source
- 40 of high voltage. The deuterons are thus projected at high speed and pass through the cathode B. The deuterons fall on a substance 13 in a sealed container 13A. The substance 13 consists, for instance, of lithium. The collision of the fast
- 45 diplogen ions with the substance 13 causes transmutation, i. e. a nuclear reaction of the deuteron with an atom of the target. The substance 13 is surrounded by a thick layer 14 containing the element which it is desired to transmute into a
- 50 radio-active element. In order to have a high efficiency, the thickness of the layer 14 has to be sufficiently great, compared with the mean free path of the neutron, to prevent escape of any of the neutrons.

55 Figure 2 shows in more detail the electrical dis-

charge tube 11 referred to in Figure 1. The tube essentially consists of a main portion 16 serving to accelerate the deuterons and an auxiliary tube IIT for initiating the flow. IIA is the anode and 15 the cathode of the auxiliary tube, deuterium being admitted thereto through the inlet 13B and being pumped away through the outlet 14A. The flow initiated by the auxiliary tube is accelerated by passage through the main tube 16 which is maintained exhausted by suction outlets 141 and 10 14<sup>2</sup>, and which has a high potential gradient, there being a million volt potential difference between the ends of the tube. The accelerated deuterons emerge through the neck  $14^3$ , of the tube 16 and collide with the substance 13 as de- 15 scribed with reference to Figure 1 of the drawings

If the substance 13 is a light element for instance lithium, then the bombardment by the accelerated deuterons results in emission of un- 20 charged particles of mass of the order of magnitude of the mass of a proton. Such uncharged nuclei i. e. neutrons, penetrate even substances containing the heavier elements without ionisation losses, and will cause the formation of radio- 25 active substances in the layer 14 exposed to them. It is to be noted that by the method so far described, the ionisation losses suffered by the deuterium nuclei are comparatively small in light elements and also that the substance to be made 30 radio-active is irradiated with neutrons i. e. uncharged nuclei, which pass through even heavy elements without ionising them. The substance 14 exposed for treatment by the neutron radiation may be in the form of an organic compound 35 for the purpose of carrying out separation of the generated radio-active element, as described more fully hereinafter.

Neutron radiation may also be produced by the action of X-rays upon an element having a dis- 40 sociable neutron at the prevailing voltage, and apparatus for carrying out this process will now be described with reference to Figure 3 of the drawings.

In Figure 3, 1 is the primary of a transformer, 45 the secondary 2 of which is connected to the junctions 3 and 4. The junction 3 is connected to the cathode 8 of the rectifier tube 5 and to the anode 7 of the rectifier tube 6. The junction 4 is connected to the cathode 9 of the rectifier 50 tube 10 and to the anode 11 of the rectifier tube 12. The cathodes 13 and 14 are connected to each other and to earth. The anodes 15 and 16 are connected at 17, and fom this point are connected to the pole 18 of the impulse generator 20, 55 the pole 19 of which is connected to earth. The impulse generator 20 is built of condensers 21, resistances 22 and spark-gap devices 23.

- The impulse generator and rectifying unit 5 shortly described above, are known components adapted to give an extremely high voltage for a fraction of a second. With such a system voltages up to 3 million volts have been obtained. The negative side of the impulse generator is con-
- 10 nected to a spark gap device 25, which in turn is connected with the cathode 26 of the discharge tube 24. The latter is built up from rings 24A of which only a few are shown in the drawings. It will, however, be understood that the rings
- 15 are continuous to enclose a space which is exhausted through the outlet 24B. The anode 27 of the tube is connected to earth and is formed by a metallic window. A body of material 28 is arranged at the external side of the window 27.
- When the impulse generator operates to pro-20 duce discharge between the cathode 26 and anode 27 of the tube 24, fast electrons penetrate the anode 27 and impinge upon the body 28. The latter when formed of Bi or Pb or some other

as heavy element, efficiently acts as an anti-cathode and hard X-rays are produced. In Figure 4 of the drawings there is shown the

lower portion of the discharge tube 24 with a device therebeneath for utilising the hard X-rays

- 30 capable of being produced with the aid of the fast electrons emerging through the anode 27 of the tube 24. The device consists of a block 34 of the element which is to be made radioactive, a block 32 of an element with a dissociable
- 35 neutron, being located therein. An aperture is formed in both the blocks 32 and 34 to allow entry of the cathode rays from the tube 24 above. The blocks 32 and 34 are also arranged to accommodate a wheel 30 and axle 35. The wheel 30
- 40 at its periphery carries a covering of tungsten or lead 31. The covering 31 acts as an anti-cathode and is cooled with water introduced along the bearing for the axle 35. The block 34 may be in the form of a cube having a length of
- 45 side of 50 cm., whilst the block 32 can also be of cube form with a side of 25 cm. For the sake of example the block 34 may be formed of iodine or arsenic or other material which lends itself to being made radio-active. The block 32 may
- 50 be of metallic beryllium. In order that an isotopic separation as described hereinafter may be performed after irradiation the material of the block 34 may be in the form of an organic compound. A voltage of 3 million volts may be used
- 55 for the discharge tube and in operation the wheel 30 is rotated so that electrons passing through the anode 27 of the tube 24 hit the rotating anticathode covering 31. When the fast electrons strike the anti-cathode, hard X-rays are pro-
- 60 duced which penetrate the beryllium block 32 and cause neutrons to be released therefrom. which neutrons then act upon the block 34.

It may be that fast electrons and hard X-rays have a similar effect upon beryllium and one may 85 therefore contemplate the making of the covering 31 of the wheel 30 from beryllium, the beryllium block 32 then being dispensed with, so that the neutrons released directly from the beryllium anti-cathode may enter and act upon the block 70 34.

It is found that when various elements are irradiated with neutrons by the process described above, practically all elements which become radio-active transmute into their own radio-

75 active isotopes, and it becomes difficult to sepa-

rate these radio-active isotopes from the remaining portion of the element unaffected. In order to achieve separation of the radio-active element from the non-radio-active part thereof the fol-lowing process may be adopted. This process is based on the fact that if a compound of an element is irradiated by neutrons, and if an atom of the element transmutes into the radio-active isotope, then this atom is freed from the compound. In accordance with the process, a compound of the element it is desired to make radioactive is chosen such that the freed radio-active isotope of the element will not interchange with the combined atoms of the element within the compound, whereby the freed isotope may be chemically separated from the irradiated compound. Very often the element whose radioactive isotope is to be isolated, can be conveniently irradiated in the form of a compound in which it is bound to carbon. Thus in the case ; of iodine compounds such as iodoform or ethyl iodide, the radio-active iodine isotope may be chemically separated from the original iodine compound in the form of free iodine. In order to protect the radio-active iodine isotope a small 25 amount of normal iodine may be dissolved in the organic iodine compound before irradiation or after irradiation but before separation.

What I claim and desire to secure by Letters Patent of the United States is:

1. The method of producing a radio-active element from a natural element by causing fast deuterons to impinge on a target containing lithium, and exposing a layer of the natural element to be transformed into a radio-active ele- 33 ment to the neutron radiation emitted by the said target.

2. The method of producing from a natural element a concentrate of a radio-active element which is isotopic with the said natural element, 40 which comprises subjecting a compound of said natural element to an irradiation which will transform some of said natural element into a radio-active isotope of said natural element, said compound of said natural element being one 45 which in the environment in which the irradiation is being carried out does not interchange atoms of said natural element bound in the compound with atoms of said natural element or its isotopes outside the compound, and separating, 50 after irradiation, from the compound said natural element and its isotopes which are outside the compound.

3. The method of producing from a natural element a concentrate of a radio-active element 55 which is isotopic with said natural element, which comprises subjecting a compound of said natural element to irradiation with neutrons which will transform some of said natural element into a radio-active isotope of said natural ele- co ment, said compound of said natural element being one which does not interchange in the environment in which the irradiation is carried out, atoms of said natural element bound in the compound with atoms of said natural element 65 or its isotopes outside the compound, and separating, after irradiation, from the compound said natural element and its isotopes which are outside the compound.

4. The method of producing from a natural 70 element a concentrate of a radio-active element which is isotopic with said natural element, which comprises irradiating with neutrons an organic compound of said natural element which will not interchange atoms of said natural ele- 75

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ment bound in the compound with atoms of said natural element or its isotopes outside the compound, and separating, after irradiation, from the compound said natural element and its isotopes outside the compound.

5. The method of producing from a natural element a concentrate of a radio-active element which is isotopic with said natural element, which comprises irradiating with neutrons a compound

- 10 which contains carbon, in which said natural element is bound to carbon and which compound will not interchange atoms of said natural element bound in the compound with atoms of said natural element or its isotopes outside the com-
- 15 pound, and separating, after irradiation, from the compound said natural element and its isotopes outside the compound.

6. The method of producing from a natural element a radio-active element which is isotopic 20 with the natural element comprising the steps

- of producing fast electrons, directing the steps ward a target adapted to produce X-rays under the impact of said electrons, exposing to the action of said X-rays an element of the class
- 25 consisting of beryllium and heavy hydrogen which produce neutron radiation under the action of said X-rays, and producing a radio-active element from a natural element by exposing the natural element to said neutron radiation.
- **30** 7. The method of producing from a natural element a radio-active element which is isotopic with the natural element comprising the steps of producing fast electrons having an energy of at least 3,000,000 volts, directing them toward a
- 35 target adapted to produce X-rays under the impact of said electrons, exposing to the action of

said X-rays an element of the class consisting of beryllium and heavy hydrogen from which neutrons are liberated by X-rays of 3,000,000volts energy, and producing a radio-active element from a natural element by exposing the 5 natural element to said neutron radiation.

8. The method of producing from a natural element a radio-active element which is isotopic with the natural element comprising the steps of producing fast electrons, directing them to-10 ward a target adapted to produce X-rays under the impact of said electrons, exposing beryllium to the action of said X-rays to produce neutron radiation, and producing a radio-active element from a natural element by exposing the natural 15 element to said neutron radiation.

9. The method of producing from a natural element a radio-active element which is isotopic with said natural element comprising the steps of producing fast electrons, directing them to-20 ward a target adapted to produce X-rays under the impact of said electrons, exposing to the action of said X-rays an element of the class consisting of beryllium and heavy hydrogen which produce neutron radiation under the action of 25 said X-rays, and irradiating by said neutron radiation a compound of said natural element which in the environment in which said irradiation is carried out will not interchange atoms of said natural element bound in the compound with 30 atoms of said natural element or its isotopes outside the compound, and separating, after irradiation from the compound said natural element and its isotopes outside the compound.

LEO SZILARD.

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The invention concerns a process and apparatus for the transmutation of elements and makes it possible to bring about such a transmutation on a large scale. The production of power by means of the heat generated in the process and the production of radio active elements on a large scale are thus made possible by the invention. The invention teaches that it is possible to produce a nuclear chain reaction and to maintain the stationary conditions in such chain reactions. It also teaches that it is possible to have explosive bodies in which an explosion is brought about at will by a sudden change in the distribution of matter.

Johed March 6

According to the invention such a chain reaction may be maintained for instance by maintaining an initial radiation of neutrons by means of one of the known methods for the production of neutron radiations (for instance by bombarding a lithium target with deuterons or by exposing beryllium to X-rays or gamma-rays) and letting these neutrons fall on a body/suitably chosen substance, shape and dimensions and thereby obtaining an increase in the number and energy of the neutrons which increase is brought about by their interaction with the said body. Thus if the proper conditions are observed the interaction of neutrons with matter can lead to the liberation of further neutrons. These newly liberated neutrons liberate again in their turn further neutrons so that we have a chain reaction in which a large number of neutrons are liberated, the total number being limited only by the dimensions of the geometrical arrangement.

-2-

As a carrier of such a chain reaction it is necessary to have an element the mass of which is sufficiently high to allow its disintegration into its parts with liberation of energy, the disintegration being ordinarily inhibited (meta stable state), the inhibition being lifted in a reaction with a neutron. An example of such a meta stable element is uranium.

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In order to be able to utilize a nuclear reaction in which an excess number of neutrons is liberated by neutrons for the maintenance of a chain reaction, it is not sufficient to measure the cross-section and other constants of the reaction, but it/also necessary to be aware of the laws which govern the neutron output of such reactions in function of the geometrical conditions. Once the general laws, the type of behavior is known, the exact dimensions can be easily determined in each particular case by actually measuring the neutron output. Such experimental adjustments can, however, only be made if the general type of behavior is known.

In the following we shall by way of example demonstrate certain general features of such a neutron chain reaction in a special case. In this special case the carrier of the chain reaction forms a spherical layer as illustrated in Fig. 1. In Fig. 1, 1 is a spherical layer which contains a carrier of the chain reaction, 2 is a neutron source which has fixed constant output of neutrons,  $\underline{r}_1$  is the inner radius of the spherical layer. In order to be able to treat the problem as a diffusion problem, we choose  $\underline{r}_1$  very much larger than the mean free path <u>a</u> of the neutrons produced in the chain reaction in the substance of

-3-

Such change is not only possible in the presence of a meta stable element. A meta stable element is an element the mass of which (packing fraction) is sufficiently high to allow its disintegration into its parts and a liberation of energy. Elements like uranium and thorium are examples of such elements. It is possible to measure the packing fraction of the elements by means of the mass spectograph and thereby determine whether an element is meta stable or not in the sense in which the word is used in this application. If, for instance, the values which were generally accepted in 1934 for helium and beryllium had been correct (and we know today that they were false, at least in their relation to each other), we have been correct in concluding from these values which were accepted in 1934 that beryllium is a meta stable element and can be disintegrated into parts with a liberation of energy, one of the parts set free in its disintegration being a neutron. If such an element which is meta stable but the disintegration of which is inhibited is exposed to neutrons, we may have a reaction in which a neutron disappears and more than one neutron is emitted. The additional neutrons would together with the number of the original neutrons continue to interact with the meta stable element thereby forming the links of a chain reaction.

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In order to be able to utilize a nuclear reaction in which an excess number of neutrons is liberated by neutrons for the maintenance of a chain reaction, it is not sufficient to measure the cross-section and other constants of the reaction, but it is also necessary to be aware of the laws which govern the neutron output of such reactions in function of the geometrical conditions. Once the general laws, the

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the chain reaction layer. The neutrons liberated in the chain reaction layer will have to make a number of collisions with the nuclei which compose the chain reaction layer, and we designate the number of collisions necessary for causing transmutations in the carrier of the chain reaction layer which lead on the average to the liberation of one additional neutron with f. We further assume that f is a large number so that we may have conditions in which the well known equations of diffusion can be applied. With these assumptions the density s of the neutron will with good approximation be given within the chain reaction layer as a function of the radius <u>r</u> by the following equation: (1)  $D \frac{d^2(rs)}{dr^2} \neq A(rs) = 0$ (1)

D is a diffusion constant which is given by

D = aW

X.h. Wis the mean velocity of the neutrons; a the mean free path for scattering collisions within the chain reaction layer; and f a number which says how many scattering collisions a neutron has to make in the chain reaction layer in order to produce on the average an additional neutron.

> A stands for the number of neutrons produced per c.c. of chain reaction layer in a second and its value is given by

A = at so that VD = alf

For a given value of  $\underline{r}_1$  there is a certain value of  $\underline{r}_2$ and accordingly a certain value of  $\underline{r}_2 - \underline{r}_1$  for which the number of neutrons per second diffusing out of the spherical layer into space becomes infinite for a finite neutron production of the source. This value of  $(\underline{r}_2 - \underline{r}_1)$  we may call the critical thickness of the chain reaction layer. If the

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thickness of the chain reaction layer is smaller than but very close to the critical thickness, the neutron output is very much larger than the neutron input and we may have 1000 or more times as many neutrons emerging from the chain reaction layer as the neutron input of the neutron source 2. The value of the critical thickness is a function of ry and of the boundary conditions for  $\underline{r} = \underline{r}_1$  and  $\underline{r} = \underline{r}_2$ . If the outer surface  $(\underline{r} = \underline{r}_2)$  of the spherical layer were to stand free in space, the density s would be 0 for the outer surface, and if there is no absorption of neutrons in the hollow sphere containing the neutron source, i.e. if the number of neutrons produced by the neutron source is equal to the number of neutrons diffusing outwards from the sphere  $\underline{r} = \underline{r}_1$ , we obtain very large values of  $\underline{r}_1 (\underline{r}_1 >> \sqrt{\frac{3}{4}})$ ) for the value of the critical thickness (2)  $L_{st} = \frac{T}{2} \left| \frac{D}{A} \right| = \frac{T}{2} \frac{a/F}{\sqrt{3}}$ 

The critical thickness for these conditions we shall call further below the standard critical thickness of the substance which composes the chain reaction layer.

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(2)

If the outer surface is covered by some material, for instance if the transmutation layer is covered by lead or immersed into water, the critical thickness is less than the standard critical thickness. This is due to the back scattering by lead or water, and, in the case of water, the fact that the neutrons are slowed down by water and their mean free path is thereby reduced plays an important role.

If the neutrons are allowed to escape out of the hollow sphere containing the neutron source in the interior of the spherical layer or if they are absorbed within this hollow sphere, the critical thickness is increased.

If all the neutrons were captured in the hollow sphere within  $\underline{r} = \underline{r}_1$ , the critical thickness would become independent of the value of  $\underline{r}_1$  and have a value exactly double the normal critical thickness provided that the neutron density  $\underline{s}$  remains 0 at the outer surface ( $\underline{r} = \underline{r}_2$ ).

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Obviously the above given diffusion equation holds only for stationary solutions, that is for solutions where the neutron density <u>s</u> is a function of <u>r</u> only and does not vary with time. Not for all boundary conditions will such stationary solutions exist. If we gradually increase the thickness of the **Artical** (layer we reach for any given boundary conditions a thickness at which the neutron outflow becomes infinite for finite neutron production of the flue neutron source. This thickness is  $\neq$  critical thickness of the arrangement. It can be calculated for every case in the following way from given boundary conditions.

The solution of the above given diffusion equation has the form of (3)  $sr = C, sin(\overline{A}, r) + C_2 cos(\overline{A}, r)$ 

For the boundary condition  $\underline{s} = 0$  for  $\underline{r} = \underline{r}_2$  the solution takes the form of (4)  $ST = C Sin[T_2 - T_1] \sqrt{\frac{D}{A}}]$ 

or (5)

s = c sin[(n-r) / JA-]

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any sphere is given by  $4\pi r^2 D \frac{ds}{dr}$ 

and it is therefore also

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To take another example, if the boundary conditions are  $\underline{s} = 0$  both for  $\underline{r}_1$  and  $\underline{r}_2$ , then a glance at equation (5) shows that  $(\underline{r}_2 - \underline{r}_1) = \overline{n} / \overline{\frac{p}{A}}'$ , i.e. twice the critical thickness, quite independent of the value of  $\underline{r}_1$ .

Figs. 2 and 3 show such a chain reaction apparatus. A neutron radiation, the initial radiation, is generated by the high voltage canal ray tube 1 (shown in greater detail in Fig. 6). This tube generates fast deuterons which strike the target 28 which contains deuterium. The neutron radiation emerging from 28 acts on the matter 3 which fills the spherical transmutation space. The composition of this matter 3 will be discussed further below and is such thet a chain reaction is released by the neutrons. The pumps 120, 121 and 122 pump a liquid, for instance water or mercury, through the pipe systems 107, 110, 111 thereby cooling the transmutation area 3 and driving the heated liquid through the boiler 126. The boiler supplies steam to a power plant. The neutrons emerging from the sphere 3 act on a layer 9 which is composed of an element xx that will trans-

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mute into a radio-active body (which is suitable for the storage of energy).

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An essentially different way of introducing the initial radiation into the chain reaction chamber is the arrangement shown in Fig. 4. 401 is the cathode ray tube described in Fig. 1. 402 is a sheet of a heavy element for instance Pb, or U in which penetrating radiation (hard X-rays) is generated with an extremely good efficiency if the electrons have a voltage over one million volts. This efficiency increases very rapidly with the voltage, and is much higher than it could be expected from the experience based on ordinary X-ray work. The thickness of the sheet 402 is such as to enable the generated penetrating radiation to penetrate through this sheet and act on the transmutation chamber 106 (in Fig. 27. Nevertheless the sheet can be sufficiently thick to utilize more than half of the energy of the cathode rays. The X-rays emerging from sheet 402 penetrate the layer 3 and can liberate neutrons either from the layer 3 or from a substance 407 placed in the interior of the layer 3). For instance, if we have beryllium present in 403 or in 3 neutrons will be liberated by X-rays. These neutrons can then maintain a chain reaction as discussed further above and further below. The advantage of using X-rays as an initial radiation is the following: The X-rays penetrate through a perfectly closed layer 3 into the interior of the layer and therefore a leak of neutrons from the interior can be avoided. This is specially important if we have to deal with a neutron chain in which no multiplicator action is involved. In such cases X-rays may be used with

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advantage as initial radiation especially in view of the unexpectedly large efficiency of the X-ray production by means of fast electrons acting on heavy elements.

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We wish to discuss now certain features which arise when the above teachings of this application are applied to uranium. If uranium is exposed to neutrons of a few million volts energy or to neutrons slowed down by water to a few volts of energy or less, uranium emits neutrons in two different ways. This can be demonstrated with particular ease if uranium is exposed to slow neutrons and the slow neutron stream hitting the uranium is suddenly stopped. One then finds that uranium emits neutrons after the stoppage of the slow neutron stream for another few, perhaps ten to fifteen, seconds. This delayed neutron emission is weak, i.e. there are less than perhaps thirty neutrons emitted in this way per incident neutron which disappears in reacting with the uranium. No chain reactions could be based on this delayed neutron emission alone. On the other hand, it is easily demonstrated that uranium emits a large number of neutrons while it is exposed to the slow neutron stream and the number of neutrons which are thus instantaneously (within a fraction of a second) emitted from uranium per number of incident neutrons (which are captured in the reaction with uranium) is larger than one. This fact makes it possible to use uranium as the carrier of a chain reaction. The neutrons which are emitted from uranium during the irradiation with slow neutrons can be distinguished from the incident slow neutrons by virtue of the fact that their velocity is much higher and ranges somewhere between a few ten thousand volts to perhaps about

a million volts or so.

In spite of its small value the delayed neutron emission is not without significance. In view of its existence we have - in order to be more precise - to distinguish between the critical thickness for the total neutron emission (instantaneous and delayed emission together).

It is possible to use uranium in the chain reaction layer not only as uranium metal or as uranium oxide but also it is possible to use either of these mixed with a hydrogen containing substance such as for instance water and it is possible to use such a large concentration of the hydrogen containing substance that most of the neutrons are slowed down to a few volts before they react with the uranium in releasing an additional number of neutrons (releasing more than one neutron on the average for every neutron captured by uranium). That the use of hydrogen containing substances in such concentrations should be possible is very surprising and unexpected and is in contradiction with the generally accepted value of the capture cross-section of uranium for neutrons slowed down by water to a few volts velocity or less. The total cross-section of uranium has been assumed on the basis of published measurements to be above 40 x  $10^{-24}$ , i.e. the capture cross-section well above 30. On the other hand, the cross-section for that transmutation of uranium which yields on the average neutrons in excess of one but certainly not in excess of 8 per transmutation is known to be less than 3. In these circumstances most of the neutrons which are slowed down to a few volts or less will be captured by uranium without causing a transmutation of the type which yields neutrons. Accordingly one would expect that

less than one neutron will be emitted on the average for every neutron captured by standard uranium. (By "standard uranium" we mean uranium in which the relative abundance of the two isotopes 135 and 138 has the ratio which one finds in uranium that occurs in nature, i.e. about 1:139.) The only possible explanation is that for some reason or other an error has been made by those who previously measured the total cross-section or uranium to be about 43 x 10<sup>-1</sup> though it is not possible to state through what mistake such error occurred.

The mixing of uranium with a hydrogen containing substance to form the chain reaction layer has the advantage greatly to reduce the critical thickness and also the amount of uranium required. The uranium or uranium oxide can either be mixed with the hydrogen containing substance or the chain reaction layer may be built up from alternating layers of uranium and the hydrogen containing substance. As a hydrogen containing substance water, paraffin wax or calcium hydride appear to be suitable. In If the hydrogen containing substance is not mixed with uranium, but alternating layers are used, the layers should be as thin as possible. In particular the thickness of the hydrogen containing layer should be as small as possible, and if paraffin, water, or a hydrogen containing substance of about the same hydrogen concentration is used, the thickness of the layer should not exceed about 7 mm.

It is important that the ratio of the number of grams of hydrogen to the number of grams of uranium in the chain reaction layer shall not exceed a certain critical value as otherwise no chain reaction can be maintained for any thickness of the layer which contains the mixture.

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This is due to the fact that hydrogen captures slow neutrons and therefore competes with uranium. The capture crosssection of the hydrogen atom is about one-third  $x10^{-24}$ and the capture cross-section of uranium for the transmutations which lead on the average to the liberation of about one additional neutron is about 2 x  $10^{-24}$  sq. c. If these approximate values were exactly correct, no chain reaction would be possible in a layer which contains hydrogen and uranium in a ratio of 6 grams of hydrogen to 238 grams of uranium. It is certain that no chain reaction is possible in a layer which contains 20 grams of hydrogen for every 240 grams of uranium. On the other hand, it is desirable in order to keep the critical thickness low not to use too little hydrogen. A reasonable value for the ratio of hydrogen to uranium which gives good results and none too large critical thickness can be found if the

expression (7)

is plotted and a value of Xm is selected for which the expression becomes maximum. If a mixture is prepared in which the ratio of the number of hydrogen atoms to the number of uranium atoms  $N_{\rm H}/N_{\rm L}$  has a value between

 $X \bigvee \frac{1}{2-\frac{2}{2}X}$ 

which the isotope 235 has been concentrated so that its relative abundance is larger than one part in 139, a larger concentration of hydrogen can be used in the chain reaction layer. The ratio of the hydrogen to the uranium concentration can be increased by the same factor by which the relative abundance of the uranium isotope 235 has been increased. Such an increase of the relative abundance of the uranium isotope 235 can be achieved by subjecting uranium hexa fluoride to one of the known diffusing processes which lead to separation of molecules of different molecular weight.

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In such a chain reaction layer which contains such large concentrations of hydrogen as indicated above, the neutron emitted from the uranium in the chain reaction is slowed down after traveling in the chain reaction layer an average distance b from its origin. At the distance b from its origin the neutron will therefore be so show that its mean free path a for scattering is much smaller than the value b. In paraffin wax this mean free path a would be about 2-1/2 mm, and in the chain reaction layer it will be comewhat larger, i.e. by a factor k which gives the ratio of the concentration of hydrogen in paraffin to the concentration of hydrogen in the chain reaction layer. This reduction of the mean free path for scattering is due to the large scattering cross-section of hydrogen for room temperature neutrons. In addition, the neutrons which have been slowed down at an average distance b from their origin have now, being slow, a large cross-section for those transmutations of uranium which lead to the liberation of neutrons. The combined effect of the large scattering cross-section of the slow neutrons in the chain reaction layer and the large transmutation cross-section of the slow neutrons for uranium is that a neutron which is emitted by uranium and which becomes slow at the average distance <u>b</u> from its point of origin will transmute a uranium nucleus at a point which is at an average distance <u>q</u> from the point at which the neutron became slow, and the distance <u>q</u> is small compared to <u>b</u>, so that the neutron will transmute a uranium nucleus at a distance <u>b</u> not very different from <u>b</u> from the point of its origin.

In these circumstances the previously given equation no longer holds, and the critical thickness is approximately given in the following way: For the stationary state the neutron density within the spherical chain reaction layer obeys now the equation: ,

obeys now the equation. (9)  $(3)^{\circ} d(7) + \frac{6}{47^{\circ}} (7)$ , (7)In this equation  $w_0$  is the probability for a slowed down neutron to cause a transmutation of uranium in which the slow neutron disappears and no fast neutron is emitted;  $w_1$  is the probability for a slowed down neutron to cause a transmutation of uranium in which the slow neutron disappears and one fast neutron is emitted;  $w_2$  is the probability for a slowed down neutron to cause a transmutation of uranium in which the slow neutron to cause a transmutation of uranium in which the slow neutron disappears and two fast neutrons are emitted.

For the special case:  $\underline{W}_2 = 1$ ;  $\underline{W}_1 = 0$ ;  $\underline{W}_0 = 0$ , the above equation gives

 $(\bar{b})^n \frac{d(rs)}{dn-} + 3(rs) = 0$ 10

 $(11) \qquad L_{st} = \frac{T}{3} \frac{6}{3(w_1 - 2w_2)}^{1/2}$ 10= 1 8 12 (12)

in the special case.

Obviously, the above diffusion equation presupposes for its validity a small value of  $\underline{w}_2$ , but even for large values of w it gives at least the order of magnitude for the critical thickness.

The critical thickness will in practice always be determined empirically for instance in the following way: a neutron source is surrounded by the chain reaction layer of an approximately correct thickness which is safely below the critical thickness. The radiations emitted from the chain reaction layer while exposed to this neutron source are observed by means of an ionization chamber. Then the thickness of the chain reaction layer is brought closer to the critical thickness by gradually increasing either the quantity of uranium or the quantity of hydrogen containing substances mixed with the uranium. The amount of ionizing radiation which is emitted is again observed and the thickness of the chain reaction layer is again brought closer to the critical thickness in the same way as before. In this way, by observing the increase of the emitted radiation as a function of the increasing effective thickness of the chain reaction layer the critical thickness can be extrapolated from the observed curve by plotting the intensity of the emitted neutron radiation against the effective thickness of the chain reaction layer. Instead of an ionization chamber which registers

the neutron intensity by means of recoil ions in the gas of the chamber, induced activity caused by the neutrons can be used as a measure of the radiation intensity.

# Variation of Critical Thickness

If slow neutrons are used the critical thickness can be increased by having a slow neutron absorber within the hollow sphere in the center of the spherical arrangement. If the inner radius of the spherical shell of the chain reaction layer is much larger than the critical thickness (to be accurate we have said the standard critical thickness given by the above formulas), and if all slow neutrons are absorbed, for instance by a cadmium layer covering the inner surface of the spherical chain reaction layer, the critical thickness of the arrangement is increased. By removing such absorbing matter from the inside of the chain reaction layer, the critical thickness may be reduced below the actual thickness, and thus an explosion may be brought about. The explosion will be all the more violent the more quickly the absorbing substance is removed. A similar increase in the critical thickness of a spherically symmetrical chain reaction layer can be brought about by removing a section of the layer and thereby producing an opening through which the neutrons can escape. For instance a conical section corresponding to a few percent of the spherical chain reaction layer can be so arranged as to be easily moved out of its place and replaced, and thereby the critical thickness may be reduced or increased.

Explosions may be brought about by a variation of the critical thickness for the purpose of creating a large amount of radio active elements. In such a case it is

desirable to have mild explosions. Mild explosions can be brought about by slowly changing the space distribution of matter and also by arranging layers so that when the explosion sets in there shall be no rapid removal of substances which capture neutrons and thereby reduce the critical thickness. For instance, if the chain reaction layer is based on fast neutrons and we have a substance in the hollow sphere in the center of the chain reaction layer which will slow down neutrons and absorb the slow neutrons, for instance a solution of a boron salt in water, the arrangement may be such that there shall be no easy outflow for the water from the inside of the spherical chain reaction layer towards the outside. On the contrary, if a strong destructive explosion is wanted. it can be brought about by providing for such outflow. It is advisable to have explosions which for the purpose of reducing radio active elements are so arranged that the explosions shall take place in the middle of a large water tank or below water. After the explosion the scattered radio active material can then be collected from the water.

# Regulation

As we have seen, the ratio of neutron input to neutron output becomes infinite for the critical thickness of the arrangement. The neutron input is in practice limited by the accuracy of the arrangement, since the thickness of the chain reaction layer must be extremely close to the critical thickness, and yet must remain below it in order to avoid an explosion. Fortunately, it is possible to overcome this difficulty by reason of the

### following fact:

In reality we have to deal not with one critical thickness only, but with two different critical thicknesses which we shall call the instantaneous critical thickness and the delayed critical thickness. They arise by virtue of the fact that, while the bulk of neutrons is emitted instantaneously when uranium is transmuted by neutrons. there is also a delayed emission of neutrons, the delay being of the order of magnitude of a few seconds. If the thickness of the chain reaction layer is larger than the delayed critical thickness, but smaller than the instantaneous critical thickness, the neutron output increases to infinity, but does not increase too rapidly. This makes it possible that by moving an object which forms part of the arrangement and which has an influence on the critical thicknesses (for instance, by having a slow neutron absorber in the interior of the hollow sphere of the spherical chain reaction layer, and by partially withdrawing it from there, we can reduce the critical thickness, and in a similar way we can also increase it by the opposite movement), we can vary the critical thickness in time. We shall call objects which are used in this way regulator objects, and according to our invention the neutron output can be kept very high without risking an explosion by moving the regulator object in such a way that part of the time the critical thickness for delayed emission should be below, and part of the time it should be above, the real thickness of the spherical layer. It is only necessary for safe functioning to have an instrument which is sensitive to the emitted radiation or the temperature of some part of the chain reaction

layer, and this instrument can control the position of the regulator object. Obviously, in order to have stable functioning, the regulator object will have to be moved in direction of an increase of the critical thickness with increasing neutron radiation, and it has to be moved in the opposite way with decreasing neutron radiation. While the thickness of the chain reaction layer will still have to be accurately chosen, since it has to be within narrow limits, i.e. between the critical thickness for instantaneous neutron emission and the critical thickness for delayed neutron emission, the latter being only slightly larger than the former. Yet the above described regulation makes it possible to get a very much higher neutron output without reaching an explosion.

The critical thickness for delayed emission could also be conveniently called the total critical thickness because it corresponds to the total neutron emission, both instantaneous and delayed. It can be easily determined empirically by varying the thickness of the chain reaction layer and observing for each thickness the emitted neutron radiation as a function of time. Below the critical thickness for delayed emission the neutron radiation is a function of time which resembles a growth curve in the field of radio activity, i.e. it approaches an upper limit practically reaching saturation after some time. Above the critical thickness for delayed emission, but below the critical thickness for instantaneous emission, the observed neutron emission increases more and more rapidly with time, and the arrangement has quickly to be changed in order to avoid overheating. The value of the critical thickness for

delayed emission is reached when one type of curve goes over into the other, and at the critical thickness itself the neutron intensity as a function of time is a straight line.

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We wish now to insert a description of Figs. 5, 6 and 7 to which we have referred in the preceding part of the specification.

Fig. 5 shows an example of a suitable arrangement. 11 is an electrical discharge tube ejecting a beam 12 of fast diplogen ions.\* The ions fall on a substance 13 consisting of for instance gaseous diplogen or a diplogen compound or lithium, causing transmutation, i.e. a nuclear reaction of the diplogen ion with an atom of the target. The substance 13 is surrounded by a layer 14 containing the element which we wish to transmute into a radio active element. In order to have a good efficiency, the thickness of the layer 14 has to be sufficiently large, compared with the mean free path of the neutron for this transmutation.

Fig. 6 shows the electrical discharge tube referred to in Fig. 5. It is a high voltage positive ray tube. There is an auxiliary positive ray tube on top of the high voltage tube. 11 is the anode, 15 the cathode of this auxiliary tube. Diplogen is admitted through the tube 13 and pumped away through

Fig. 7 shows an arrangement suitable for the production of hard X-rays. 1 is the primary of a transformer, the secondary 2 of which is connected to the points 3 and 4. 3 is connected to the cathode 8 of the rectifier tube 5 and to the anode 7 of the rectifier tube 6. Point 4 is connected to the cathode 9 of the rectifier tube 10 and to the anode 11 of the rectifier tube 12. The cathodes 13 and 14 are connected to each other and to the earth. The anodes 15 and 16 are connected to point 17, and this point

also called diplons or deutons

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is connected to the pole 18 of the impulse generator 20, the pole 19 of which is connected to earth. The impulse generator 20 is built of condensers 21, resistances 22 and spark gaps 23.

This impulse generator is adapted to produce intermittent voltage up to 10 million volts, transmitted to the discharge tube 24 through the spark gap 25. 26 is the cathode of the discharge tube, the anode 27 of which is connected to the earth. The fast electrons emerge through the metal window 27 (which is the anode as well) and hit a body 28. This body is used as an anticathode and yields hard X-rays with very good efficiency if it is built of Bi, Pb or some other heavy element.

## I CLAIM:

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1. Production of radio active elements comprising the step of maintaining a neutron radiation in a layer containing uranium, the thickness of the said layer being slightly below the critical thickness.

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2. Production of radio active elements comprising the step of maintaining a neutron radiation in a layer containing uranium, the thickness of the said layer being slightly below the critical thickness, and the step of separating at intervals from the layer the radio active elements produced in the layer.

3. Production of radio active elements comprising the step of maintaining a neutron radiation in a layer containing uranium, the thickness of the said layer being slightly below the critical thickness, and exposing another element which can be made radio active by means of slow neutrons to the radiation of the chain reaction layer.

4. Production of radio active elements according to claims 1 to 3 comprising the step of maintaining a neutron radiation in a layer containing uranium or uranium oxide, the thickness of the layer being chosen so as to conform with equations (1) and for respectively using to fully values for the mean free path corresponding to a cross-

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5. Production of radio active elements according to claim 4, the layer being composed of metallic uranium and having a thickness of about 30 to 70 centimeters for an inner radius  $\underline{r}_1$  about 10 centimeters. 6. Production of radio active elements according to claim 5, the spherical chain reaction layer of metallic uranium being surrounded with a spherical layer of metallic bismuth the thickness of which bismuth layer exceeds 50 centimeters and the thickness of the uranium layer being below the values given in claim 5, the thickness of uranium being reduced to about half if very large amounts of bismuth are used.

7. Production of radio active elements according to claims 1 to 3 in which a hydrogen containing substance such as for instance paraffin wax or water is mixed with uranium, the number of gram atoms of hydrogen having the ratio to the number of gram atoms of uranium given by equation (8).

8. Production of radio active elements according to claim 7, the chain reaction layer being surrounded with a spherical layer of bismuth metal exceeding in thickness 50 centimeters.

9. Production of radio active elements according to claim 7, the spherical chain reaction layer being immersed in water or another similar hydrogen containing substance for instance paraffin wax.

10. Production of radio active elements according to claim 1 or claim 7 in which the uranium used has a different relative abundance of the isotopes 235 and 238 than standard uranium, the relative abundance of uranium 235 being increased by a factor of 3 or more.

11. Production of radio active elements comprising the step of increasing the critical thickness of an arrangement comprising a layer of uranium, the said increase being effected by changing the distribution of matter within or without the chain reaction layer and the increase carried to the point where the critical thickness of the arrangement is exceeded so that a chain reaction leading to an explosion takes place.

12. Production of radio active elements according to claim 11 in which the process described in claim 11 is carried out in a place surrounded by water or below water and the radio active elements produced through the explosion and scattered in the water are collected. I CLAIM:

1. Production of radio active elements comprising the step of maintaining a neutron radiation in a layer containing uranium, the thickness of the said layer being slightly below the critical thickness.

2. Production of radio active elements comprising the step of maintaining a neutron radiation in a layer containing uranium, the thickness of the said layer being slightly below the critical thickness, and the step of separating at intervals from the layer the radio active elements produced in the layer.

3. Production of radio active elements comprising the step of maintaining a neutron radiation in a layer containing uranium, the thickness of the said layer being slightly below the critical thickness, and exposing another element which can be made radio active by means of slow neutrons to the radiation of the chain reaction layer.

4. Production of radio active elements according to claims 1 to 3 comprising the step of maintaining a neutron radiation in a layer containing uranium or uranium oxide, the thickness of the layer being chosen so as to conform with equation (1) and given by equation (5) according to the prescription (6) respectively using values for the mean free path corresponding to a cross-section of uranium of about 6 x  $10^{-24}$  and oxygen of about 3 x  $10^{-24}$ and a value of <u>f</u> of about 60.

5. Production of radio active elements according to claim 4, the layer being composed of metallic urraium and having a thickness of about 30 to 70 centifor an inner radius  $\underline{r}_1$  about 10 centimeters. 6. Production of radio active elements according to claim 5, the spherical chain reaction layer of metallic uranium being surrounded with a spherical layer of metallic bismuth the thickness of which bismuth layer exceeds 50 centimeters and the thickness of the uranium layer being below the values given in claim 5, the thickness of uranium being reduced to about half if very large amounts of bismuth are used.

7. Production of radio active elements according to claims 1 to 3 in which a hydrogen containing substance, such as for instance paraffin wax or water, is mixed with uranium, the number of gram atoms of hydrogen having the ratio to the number of gram atoms of uranium given by equation (8)

8. Production of radio active elements according to claim 7, the chain reaction layer being surrounded with a spherical layer of bismuth metal exceeding in thickness 50 centimeters.

9. Production of radio active elements according to claim 7, the spherical chain reaction layer being immersed in water or another similar hydrogen containing substance, for instance paraffin wax.

10. Production of radio active elements according to claim 1 or claim 7 in which the uranium used has a different relative abundance of the isotopes 235 and 238 than standard uranium, the relative abundance of uranium 235 being increased by a factor of 3 or more.

11. Production of radio active elements comprising the step of increasing the critical thickness of an arrangement comprising a layer of uranium, the said increase being effected by changing the distribution of matter within or without the chain reaction layer and the increase carried to the point where the critical thickness of the arrangement is exceeded so that a chain reaction leading to an explosion takes place.

12. Production of radio active elements according to claim 11 in which the process described in claim 11 is carried out in a place surrounded by water or below water and the radio active elements produced through the explosion and scattered in the water are collected.

13. Production of power comprising the step of maintaining a neutron radiation in a layer containing uranium, the thickness of the said layer being slightly below the critical thickness.

14. Production of power according to claim 13 comprising the step of maintaining a neutron radiation in a layer containing uranium or uranium oxide, the thickness of the layer being chosen so as to conform with equation (1) and given by equation (5) according to the prescription (6) respectively using values for the mean free path corresponding to a cross-section of uranium of about 6 x  $10^{-24}$ and oxygen of about 3 x  $10^{-24}$  and a value of <u>f</u> of about 60.

15. Production of power according to claim 14, the layer being composed of metallic uranium and having a thickness of about 30 to 70 centimeters for an inner radius  $\underline{r}_1$  about 10 centimeters.

16. Production of power according to claim 15, the spherical chain reaction layer of metallic uranium being surrounded with a spherical layer of metallic bismuth the thickness of which bismuth layer exceeds 50 centimeters and the thickness of the uranium layer being below the values given in claim 5, the thickness of uranium being reduced to about half if very large amounts of bismuth are used.

17. Production of power according to claim 13 in which a hydrogen containing substance, such as for instance paraffin wax or water, is mixed with uranium, the number of gram atoms of hydrogen having the ratio to the number of gram atoms of uranium given by equation (8).

18. Production of power according to claim 17, the chain reaction layer being surrounded with a spherical layer of bismuth metal exceeding in thickness 50 centimeters.

19. Production of power according to claim 17, the spherical chain reaction layer being immersed in water or another similar hydrogen containing substance, for instance paraffin wax.

20. Production of power according to claim 12 or claim 17 in which the uranium used has a different relative abundance of the isotopes 235 and 238 than standard uranium, the relative abundance of uranium 235 being increased by a factor of 3 or more.

21. Production of power comprising the step of increasing the critical thickness of an arrangement comprising a layer of uranium, the said increase being effected by changing the distribution of matter within or without the chain reaction layer and the increase carried to the point where the critical thickness of the arrangement is exceeded so that a chain reaction leading to an explosion takes place. If a uranium is used which contains a high percentage of uranium 235 it is of advantage to use this uranium either mixed with a hydrogen containing substance, or to have the chain reaction layer built up from intermittent layers of uranium and the hydrogen containing substance. As a hydrogen containing substance water, paraffin, or calcium hydride appear to be suitable. The ratio of the amount of hydrogen to the amount of uranium may be anything up to about 10 grams of hydrogen for 235 grams of uranium. This ratio would roughly correspond to 100 cc paraffin wax for every 10 cc of solid uranium or about 20 cc of uranium powder.

If uranium is used in the form of an oxide the volume of uranium will be larger corresponding to the smaller density of the oxide. If the hydrogen containing substance is not mixed with the uranium, but if alternative layers of the two are used, the layers should be as thin as possible. If paraffin or a hydrogen containing substance of similar density is used the thickness of the paraffin layers should be as small as possible and should not exceed say 7 mm.

If uranium which contains a high percentage of uranium 235 is used it is of advantage to slow down the neutrons by using hydrogen containing substances, since these hydrogen containing substances reduce the mean free path for scattering in the chain reaction layer and thereby make it possible greatly to reduce the critical thickness of the chain reaction layer. This reduction of the mean free path is due to the

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high scattering cross-section of hydrogen. In addition to the decreased mean free path there is also a decrease in the number of collisions which a neutron has to make with uranium nuclei in order to produce on the average one transmutation of the uranium nucleus which leads to the liberation of neutrons. This is simply due to the fact that the transmutation of uranium 235 has a larger cross-section for slow neutrons than for fast neutrons. Practically every collision of a neutron slowed down to room temperature with a uranium nucleus leads to a transmutation process. Under these extreme circumstances the critical thickness is no longer given by the formula which we have previously described and which was derived without regard of the fact that the mean free path of the neutrons for scattering may strongly decrease with decreasing velocity.

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It is in these circumstances sometimes convenient to use compounds of heavy hydrogen, such as for instance heavy water or heavy paraffin wax or heavy calcium hydride instead of similar compounds of light hydrogen. The ratio of the amount of heavy hydrogen to the amount of uranium 235 may be

If alternative layers of the heavy hydrogen compounds and uranium are used the thickness of the layers of the heavy hydrogen compound, such as heavy paraffin wax may be anything up to ... SPECIAL DELIVERY

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March 3rd, 1939

#### REGULATION.

As we have seen, the ratio of neutron input to neutron output becomes infinite for the critical thickness of the arrangement. The neutron input is in practice limited by the accuracy of the arrangement, since the thickness of the chain reaction layer must be extremely close to the critical thickness, and yet must remain below it in order to avoid an explosion. Fortunately, it is possible to overcome this difficulty by reason of the following fact:

In reality we have to deal not with one critical thickness only, but with two/critical thicknesses which we shall call the instantaneous critical thickness and the delayed critical thickness. The arise by virtue of the fact that, while the bulk of neutrons is emitted instantaneously when uranium is transmuted by neutrons, there is also a delayed emission of neutrons, the delay being of the order of magnitude of a few seconds. If the thickness of the chain reaction layer is larger than the delayed critical thickness, but smaller than the instantaneous critical thickness, the neutron output increases to infinity, but does not increase too rapidly. This makes it possible that by moving an object which forms part of the arrangement and which has an influence on the critical thicknesses (for instance, by having a slow neutron absorber in the interior of the hollow sphere of the spherical chain reaction layer. and by partially withdrawing it from there, we can reduce the critical thickness, and in a similar way we can also increase it by the opposite movement), we can vary the critical thickness in time.

We shall call objects which are used in this way regulator objects, and according to our invention the neutron output can be kept very high by moving the regulator object in such a way that part of the time the critical thickness the critical thickness for delayed emission should be below, and part of the time it should be above, the real thickness of the spherical layer. It is only necessary for safe functioning to have an instrument which is sensitive to the emitted radiation or the temperature of some part of the chain reaction layer, and this instrument can control the position of the regulator object. Obviously, in order to have stable functioning, the regulator object will have to be moved in direction of an increase of the critical thickness with increasing neutron radiation, and it has to be moved in the opposite way with decreasing neutron radiation. While the thickness of the chain reaction layer will still have to be accurately chosen, since it has to be within narrow limits, i.e. between the critical thickness for instantaneous neutron emission and the critical thickness for delayed neutron emission, the latter being only slightly larger than the former. Yet the above described regulation makes it possible to get a very much higher neutron output without reaching an explosion.

The critical thickness for delayed emission could also be conveniently called the total critical thickness because it corresponds to the total neutron emission, both instantaneous and delayed. It can be easily determined emperically by varying the thickness of the chain reaction layer and observing for each thickness the emitted neutron radiation as a function of time. Below the

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critical thickness for delayed emission the neutron radiation is a function of time which resembles a growth curve in the field of radioactivity, i.e. it approaches an upper limit practically reaching saturation after some time. Above the critical thickness for delayed emission, but below the critical thickness for instantaneous emission, the observed neutron emission increases more and more rapidly with time, and the arrangement has quickly to be changed in order to avoid overheating. The value of the critical thickness for delayed emission is reached when one type of curve goes over into the other, and at the critical thickness itself the neutron intensity as a function of time is a straight line.

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According to the present invention a nuclear chain reaction can be maintained in a mass of substance so arranged that neutrons emitted from uranium are slowed down by carbon if the neutron source is for instance placed in the center of a large sphere of carbon with which a certain amount of uranium oxide is intermixed. The neutrons emitted by the source are slowed down by collisions with carbon muchei to thermal velocities and are then absorbed by uranium. This absorption process leads to the emission of more than one neutrons per absorbed thermal neutron, and of these neutrons emitted by uranium a small fraction may escape out of the sphere, but the rest will be slowed down by carbon to thermal velocities and will be absorbed by uranium leading again to the liberation of neutrons. A homogenous mixture of uranium oxide and carbon may for instance be used. In order to have a large ratio of neutron output to neutron input the radius of the sphere must be very close to the critical radius. This critical radius will be chiefly determined by the density of the carbon (the concentration of the carbon) and will be proportionate to this density. For a density of 2 gm per cc the critical radius will be about The conchetration of the uranium must not be too large, otherwise the neutrons are absorbed by uranium before they are slowed down to the thermal region and the chain reaction becomes impossible. On the other hand, if the uranium concentration is made too small the thermal neutrons will diffuse out of the carbon sphere before they are absorbed by uranium. A reasonable value for the ratio of the concentrations of uranium to carbon expressed in gm per cm<sup>2</sup> is

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high scattering cross-section of hydrogen. In addition to the an marcyla in the decreased mean free path there is also a decrease in the numaff agebone course from what inferors realise of ber of collisions which a neutron has to make with uranium nuclei in order to produce on the average one transmutation of the uranium nucleus which leads to the liberation of neutin oreage is rons. This, is simply due to the fact that the transmutation of which that the uranium 205 has a larger cross-section for slow neutrons than for fast neutrons. Practically every collision of a neutron Tuets slowed down to room temperature with a uranium nucleus leads to a transmutation process. Under these extreme circumstances the critical thickness is no longer given by the formula which we have, previously described and which was derived without regard of the fact that the mean free path of the neutrons for scattering may strongly decrease with decreasing velocity.

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### Variation of Critical Thickness.

If slow neutrons are used the critical thickness can be increased by having a slow neutron absorber within the hollow sphere in the center of the spherical arrangement. If the inner radius of the spherical shell of the chain reaction layer is much larger than the critical thickness (to be accurate we should have said the minimum critical thickness given by the above formulas), and if all slow neutrons are absorbed, for instance by a cadmium layer covering the inner surface of the spherical chain reaction layer, the critical thickness of the arrangement is increased. By suddenfrom ly removing such absorbing matter for the inside of the chain reaction layer, the critical thickness may be reduce below the actual thickness, and thus an explosion may be brought about. The explosion will be all the more violent the more quickly the absorbing substance is removed. A similar increase in the critical thickness of a spherically symmetrical chain reaction layer can be brought about by removing a section of the layer and thereby producing an opening through which the neutrons can escape. For instance a conical section corresponding to a few % of the spherical chain reaction layer can be so arranged as to be easily moved out of its place and replaced, and thereby the critical thickness may be reduced or increased.

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## CLAIMS:

1. Production of radioactive elements comprising the step of maintaining a neutron radiation and exposing to the said neutron radiation a layer containing uranium, the thickness of the said layer being slightly below the critical thickness.

2. Production of radiaactive elements comprising the step of exposing a layer containing uranium the thickness of which is slightly below the critical thickness to a radiation which will liberate neutrons from elements contained in the said layer and maintaining the said radiation.

3. Production of radioactive elements like 1 or 2, .... and the step of separating at intervals from the layer the radioactive elements produced from uranium.

4. Production of radioactive elements like 1 or 2, and the step of exposing another element to the neutrons emitted from the uranium in the layer under the action of the said radiation, and chemically separating the radioactive element produced by the neutrons from the said element.

5. Like 1 to 4, .... a layer containing uranium which has a larger relative abundance of the isotope 235 than natural uranium...

6. Like 12 to 4, ... a layer containing uranium in which 10% the relative abundance of the isotope 235 is 10% or greater...

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7. Process for the production of uranium in which the relative abundance of the isotope 235 is increased, comprising the step of preparing a halogen compound of uranium such as for instance UF6 from a uranium compound or uranium element, the step of subjecting the said halogen compound to a diffusion process which will lead to a concentration of the isotope 235, and the step of converting the product of this diffusion process into metallic uranium of the isotope converting

8. Like 7,... a halogen compound of uranium, in particular a chlorine compound, for instance U Cl<sub>4</sub>, from a uranium compound and a sample of chlorine which consists mainly of one of the two chlorine isotopes, and which sample can for instance be obtained by a diffusion process through which one of the two Chlorine isotopes has been enriched.

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2. Production of radisactive elements comprising the step of exposing a layer containing uranium the thickness of which is slightly below the critical thickness to a radiation which will liberate neutrons from elements contained in the said layer and maintaining the said radiation.

3. Production of radioactive elements like 1 or 2, .... and the step of separating at intervals from the layer the radioactive elements produced from uranium.

4. Production of radioactive elements like 1 or 2, and the step of exposing another element to the neutrons emitted from the uranium in the layer under the action of the said radiation, and chemically separating the radioactive element produced by the neutrons from the said element.

5. Like 1 to 4, .... a layer containing uranium which has a larger relative abundance of the isotope 235 than natural uranium...

6. Like 12 to 4, ... a layer containing uranium in which the relative abundance of the isotope 235 is 10% or greater...

7. Process for the production of uranium in which the relative abundance of the isotope 235 is increased, comprising the step of preparing a halogen compound of uranium such as for instance UF6 from a uranium compound or uranium element, the step of subjecting the said halogen compound to a diffusion process which will lead to a concentration of the isotope 235, and the step of converting the product of this diffusion process into metallie uranium. If which the component of process into

8. Like 7,... a halogen compound of uranium, in particular a chlorine compound, for instance U Cl4, from a uranium compound and a sample of chlorine which consists mainly of one of the two chlorine isotopes, and which sample can for instance be obtained by a diffusion process through which one of the two allorine isotopes has been enriched.

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Native uranium, or uranium in which the relative abundance of U 235 has been artificially increased, may be used mixed with a hydrogen containing substance to form the chain reaction layer, or alternatively the chain reaction layer may be built up from alternating layers of uranium and the hydrogen containa ing substance. As **the** hydrogen containing substance water, paraffin wax or calcium hydride appear to be suitable. If the hydrogen containing substance is not mixed with uranium, but alternating layers are used, the layers should be as thin as possible. In particular the thickness of the hydrogen containing layer should be as small as possible, and if paraffin, water, or a hydrogen containing substance of about the same hydrogen concentration is used, the thickness of the layer should not exceed about 7 mm.

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In such a chain reaction layer the neutron emitted from the uranium is slowed down after traveling in the chain reaction layer an average distance b from its origin. At the distance b from its origin the neutron will therefore be so slow that its mean free path-a-for scattering is much smaller than the value b. In paraffin wax this mean free path a would be about  $2 \frac{1}{2}$  mm, and in the chain reaction layer it will be somewhat larger, i.e. by a factor k which gives the ratio of the concentration of hydrogen in paraffin to the concentration of hydrogen in the chain reaction layer. This reduction of the mean free path for scattering is due to the large scattering crosssection of hydrogen for room temperature neutrons. In addition, the neutrons which have been slowed down at an average distance  $\overline{b}$  from their origin have now, being slow, a large cross-section for those transmutations of uranium which lead to the liberation of neutrons. The combined effect of the large scattering crosssection of the slow neutrons in the chain reaction layer and the large transmutation cross-section of the slow neutrons for uranium is that a neutron which is emitted by uranium and which becomes slow at the average distance  $\overline{b}$  from its point of origin will transmute a uranium nucleus at a point which is at an average distance q from the point at which the neutron became slow, and the distance q is small compared to  $\overline{b}$ , so that the neutron will transmute a uranium nucleus at a distance  $\overline{b}$  not very different from  $\overline{b}$  from the point of its origin.

In these circumstances the previously given equation does no longer hold, and the critical thickness is approximately given in the following way. For the stationary state the neutron density within the spherical chain reaction layer obeys now the

$$\left(\overline{b}\right)^{2} \frac{d(\overline{r}s)}{dr^{2}} + \frac{6(w_{2} - w_{0})}{w_{1} + 2w_{2}} (\tau s) = 0$$

equation:

In this equation  $w_0$  is the probability for a slowed down neutron to cause a transmutation of uranium in which the slow neutron disappears and no fast neutron is emitted;  $w_1$  is the probability for a slowed down neutron to cause a transmutation of uranium in which the slow neutron disappears and one fast

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neutron is emitted;  $w_2$  is the probability for a slowed down neutron to cause a transmutation of uranium in which the slow neutron disappears and two fast neutrons are emitted.

For the special case: $w_2=1$ ;  $w_1 = 0$ ;  $w_0 = 0$ , the above equ-

ation gives  $\frac{1}{2} + \frac{1}{2} + \frac{$ 

The critical thickness is given by  $l_{0} = \frac{\pi}{2} \overline{l} \left( \frac{(w_{2} - w_{0})}{3(w_{1} + 2w_{2})} \right)^{1/2}$ 

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Obviously, the above diffusion equation presupposes for its validity a small value of  $w_2$ , but even for large values of  $w_2$  it gives at least the order of magnitude for the critical thickness.

The critical thickness will in practice always be determined empirically in the following way: a neutron source is surrounded by the chain reaction layer of an approximately correct thickness which is safely below the critical thickness. The radiations while emitted from the chain reaction layerxxxxix exposed to this neutron source are abserved by means of an ionization chamber. Then the thickness of the chain reaction layer is brought closer to the critical thickness by gradually increasing either the quantity of uranium or the quantity of hydrogen containing substances mixed with the uranium. The amount of ionizing radiation which is emitted is again observed and the thickness of the chain reaction layer is again brought closer to the critical thickness in the same way as before. In this way, by observing the increase of the emitted radiation as a function of the increasing effective thickness of the chain reaction layer the critical thickness can be extrapolated from the observed curve by plotting the intensity of the emitted neutron radiation against the effective thickness of the chain reaction layer. Instead of an ionization chamber which registers the neutron intensity by means of recoil ions in the gas of the chamber, induced activity caused by the neutrons can be used as a measure of the radiation intensity. The maximum ratio of concentration of the hydrogen containing substance to the concentration of uranium is determined by the capture cross-section of the hydrogen atom which is about  $^{1}/3 \ 10^{-24}$ cm<sup>2</sup> to the average cross-section for the emission of two neutrons of the uranium which is used. This cross-section is about  $10^{-24}$  cm<sup>2</sup>. Therefore, if native uranium is used, the number of gm Mols of uranium in the mixture must be at least  $^{1}/3$  of the number of gm Mols's of hydrogen.

If a uranium is used in which the rare isotope has been concentrated, the amount of uranium used can be smaller in ratio of the increased average cross-section for the emission of two neutrons in the transmutation of uranium.

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> In such a chain reaction layer the neutron emitted from the uranium is alowed down after traveling in the chain reaction layer an average distance b from its origin. At the distance b from its origin the neutron will therefore be so slow that its mean free path a for scattering is much smaller than the value b. In paraffin wax this mean free path a would be about 2 1/2 mm, and in the chain reaction layer it will be somewhat larger, i.e. by a factor k which gives the ratio of the concentration of hydrogen in paraffin to the concentration of hydrogen in the chain reaction layer. This reduction of the mean free path for scattering is due to the large scattering cross

section of hydrogen for room temperature neutrons. In addition, the neutrons which have been slowed down at an average distance **b** from their origin have now, being slow, a large cross-section for those transmutations of uranium which lead to the liberation of neutrons. The combined effect of the large scattering crosssection of the slow neutrons in the chain reaction layer and the large transmutation cross-section of the slow neutrons for uranium is that a neutron which is emitted by uranium and which becomes slow at the average distance **b** from its point of origin will transmute a uranium nucleus at a point which is at an average distance **q** from the point at which the neutron became slow, and the distance **q** is small compared to **b**, so that the neutron will transmute a uranium nucleus at a distance **b** not very different from **b** from the point of its origin.

In these circumstances the previously given equation does no longer hold, and the critical thickness is approximately given in the following way. For the stationary state the neutron density within the spherical chain reaction layer obeys now the equation:

$$(\overline{e})^2 \frac{d(\overline{rs})}{d\tau^2} + \frac{6(\overline{rs}-\overline{r}_0)}{r_1+2r_2}(\tau s) = 0$$

In this equation  $w_0$  is the probability for a slowed down neutron to cause a transmutation of uranium in which the slow neutron disappears and no fast neutron is emitted; probability for a slowed down neutron to cause a transmutation of uranium in which the slow neutron disappears and one fast

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For the special case:  $w_2 = 1$ ;  $w_1 = 0$ ;  $w_0 = 0$ , the above equation gives  $(2^{1/2} - 2^{2/2}) = (2^{1/2} - 2^{1/2})$ 

 $(\overline{b})^{2} \frac{\alpha^{2}(\tau s)}{\alpha \tau^{2}} + 3(\tau s) = 0$ 

The critical thickness is given by

 $l_{0} = \frac{T}{3} \frac{1}{6} \left( \frac{w_{2} - w_{0}}{3(w_{1} - 2w_{2})} \right)^{2}$ 

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Obviously, the above diffusion equation presupposes for its validity a small value of  $w_2$ , but even for large values of  $w_2$  it gives at least the order of magnitude for the critical thickness.

The critical thickness will in practice always be determined empirically in the following way: a neutron source is surrounded by the chain reaction layer of an approximately correct thickness which is safely below the critical thickness. The radiations while emitted from the chain reaction layerxxxixixix exposed to this neutron source are observed by means of an ionization chamber. Then the thickness of the chain reaction layer is brought closer to the critical thickness by gradually increasing either the quantity of uranium or the quantity of hydrogen containing substances mixed with the uranium. The amount of ionizing radiation which is emitted is again observed and the thickness of the chain reaction layer is again brought closer to the critical thickness in the same way as before. In this way, by observing the increase of the emitted radiation as a function of the increasing effective thickness of the chain reaction layer the critical thickness can be extrapolated from the observed curve by plotting the intensity of the emitted neutron radiation against the effective thickness of the chain reaction layer. Instead of an ionization chamber which registers the neutron intensity by means of recoil ions in the gas of the chamber, induced activity caused by the neutrons can be used as a measure of the radiation intensity.

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