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PATENTS & DESIGNS ACTS 1907 to 1932

PROVISIONAL SPECIFICATION

IMPROVEMENTS IN OR RELATING TO THE TRANSMUTATION OF CHEMICALS ELEMENTS.

I LEO SZILARD, a citizen of Germany and Subject of Hungary c/o Claremont Haynes & Co., of Vernon House, Bloomsbury Square, London, W. C. l. do hereby declare the nature of this invention to be as follows:-

This invention has for its object the production of radio active bodies the storage of energy through the production of such bodies and the liberation of nuclear energy for power production and other purposes through nuclear transmutation.

In accordance with the present invention nuclear transmutation leading to the liberation of neutrons and of energy may be brought about by maintaining a chain reaction in which particles which carry no positive charge and the mass of which is approximately equal to the proton mass or a multiple thereof form the links of the chain. I shall call such particles in this specification "effecient particles".

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Ap way of bringing about effeciently transmutation processes is to build up transmutation areas choosing the composition and the bulk of the material so as to make chain reactions effecient and possible, the links of the chain being "effecient particles".

One example is the following. The chain transmutation contains an element "C", and this element is so chosen that an efficient particle "X" when reacting with "C" may produce an efficient particle "y", and the efficient particle "y" when reacting with "C" may produce either an efficient particle "x" or another efficient particle which in its turn is directly or indirectly when reacting with "C" capable of producing "x". The bulk of the transmutation area, on the other hand, must be such that the linear dimensions of the area should sufficiently exceed the mean free path between two successive transmutations within the chain. For long chains composed of, say, 100 links the linear dimensions must be about ten times the mean free path.

I shall call a chain reaction in which two efficient particles of different mass number alternate a "doublet chain" An example for a doublet chain which is a neutron chain would be the following reaction, which might be set up in a mixture of a "neutron reducer element" (like Lithium (6) or Boron (10) or preferably some heavy "reducer" element) and a "neutron converter element" which yields n(2) when bombarded by n(1). An example

for such a chain in which carbon acts as reducer and
Beryllium acts as converter would be the following:
C (12)+n (2) = C(13)+n(1) ("Be(8)" need not mean an existing element, it may break up
Be (9)+n (1) = "Be(8)"+n(2) spontaneously).

One can very much increase the efficiency of the hitherto mentioned neutron chain reactions by having a "neutron multiplicator" "O" mixed with the elements which take part in the chain reaction. A neutron multiplicator is an element which either splits up n(2) into n(1) + n(1) or an element which yields additional neutrons for instance n(1) when bombarded by n(1). A multiplicator naed not be a metastable element. Beryllium may be a suitable multiplicator.

Be(9) + n(1) = "Be(8)" + n(1) + n(1)

An efficient particle disappears (and a chain is therefore interrupted if this happens in a chain reaction), if a neutron reacts with a nucleus in such a way that the neutron disappears and a positive particle for instance a proton or an alpha particle is emitted. I can suppress the production of a positive particle when bombarding the element by neutrons by choosing the element and the neutron energy so that the positive particle, the creation of which has a potential possibility, should not have sufficient energy at its disposal to penetrate in the inverse process the nucleus of that element. In order to avoid such an occurrence in my chain reactions I shall use

as reducers, converters and multiplicators the heaviest elements which are otherwise satisfactory.

In the accompanying drawings figures 1 and 2 show an example for utilising neutron chains for power production and 101 is a high voltage the generation of radio-active bodies. positive ray tube generating fast light ions like diplons or Helium ions which cause by striking diplogen or Beryllium in 102 the emission of a penetrating radiation (neutrons). The radiation emerging from 102 acts on the material 103 which forms This material is such that a chain rea sphere around 102. action, preferably accompanied by the action of a multiplicator is released. For instance one can have a sphere 103 the dimensions of which are so chosen that the energy liberated in it should be a multiple of the energy input. 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 103 and driving the heated liquid through the boiler 126. The boiler supplies steam to a power plant. The neutrons emergying from the sphere 103 act on a layer 104 which is composed of an element "T" that will transmute into a radio-active The element body which is suitable for the storage of energy. "T" need not be present as a free element, but can preferably be present in the form of a compound soluble in water; that makes

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it easier to separate the radio active bodies formed in the d d d bodies formed in the process. A third bayer 105 contains an element "V" that will absorb the neutrons /n(1)/ under liberation of energy (Li). 106 is a heat insulating layer.

Dated this 28th

day of June 1934

Vernon House, Sicilian Avenue, Bloomsbury Square W.C.l.

Applicant's Solicitors

IMPROVEMENTS IN OR RELATING TO THE TRANSMUTATION OF CHEMICAL ELEMENTS.

LEO SZILARD ESQ.,

PROVISIONAL SPECIFICATION

CLAREMONT HAYNES & CO., Vernon House, Sicilian Avenue, Bloomsbury Square, W. C. 1. Transmutation of chemical elements by efficient particles. The invention concerns the production of power, the storage of power and the production of radioactive bodies through processes in which neutrons of simple or multiple mass, or negative nuclei are generated. The following symbols will be used for negative nuclei: H(1) for the negative proton, H(2) for the negative diplon; similarly H(3), He(3), He(4) ... for negative nuclei the mass and charge of which corresponds to the positive nuclei H(3), Elements the mass number of which is He(3), He(4). larger than twice the atomic number will be called "overloaded". Neutrons of simple or multiple mass are also considered to be "overloaded" elements. Negative nuclei may be generated by shooting "overloaded" atoms for instance on "overloaded" elements. In order to determine which element yields negative nuclei when bombarded by "overloaded" particles one has to investigate each element by means of a Wilson cloud chamber. If a magnetic field is applied to the said chamber the curveture of the tracks generated by the bombarded element will show if negative nuclei are present in the radiation of the element under investigation. If matter is radiated by negative nuclei these can be captured by the positive nuclei without necessarily falling at once into the positive nucleus, i.e. the binding energy can possibly be small at least for a certain period of time after the capture. Bombardment of Hydrogen, H(1), Diplogen H(2), Triplogen H(3) by negative protons may lead to the generation of neutrons in which the positive and negative nucleus is possibly only loosely bound for some time after the generation. For instance the binding energy

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between a positive and negative proton may at first be between ten and twenty thousand volts. Neutrone in which the nuclei are loosely bound will be called subsequently pseudo-neutrons and the symbols n(2), m(3) and n(4) to distinguish them from the real neutrons of the same mass number for which we shall write p(2), n(3) and n(4). Such pseudo-neutrons would suffer energy losses if they travel through matter at a given speed which are large as compared to the energy losses for real neutrons at the same speed. While positive nuclei do not cause transmutation if they have reached the end of their range this may be the case for may be the case negative protons and other negative muchei and for such nuclei which contain instably (loosely) bound negative nuclei, for instance pseudo-neutrons. They may be, therefore, equally efficient as real neutrons, the range of which is very large and may therefore be used in the processes and in the apparatus which is subject to this invention. We shall call them "efficient particles" and also under which name we understand neutrons, negati snd pseudo-nuclei (pseudo-nuclei are pseudo-neutrons or positive nuclei which contain loosely bound negative nuclei and therefore can equise transmutation even when at the very end of their range).

The preceding part of the description serves only to define the scope of the word "efficient particle". The following chapters deal with the invention itself which concern methods and apparatus for the production of power, for the storage of power and for the production of radioactive bodies by means of chain reactions the links of the chain being "efficient particles". 1. We choose an element "C" for our process so that its mass (packing fraction) should be sufficiently

If Neutrons are generated by bombarding a light element in the transmutation chamber 20 with Diplogen, Triplogen, or Helium ions accelerated by electric fields as indicated in Fig. 1 by means of a high voltage discharge tube as described by Cockroft and Welton, one can get strong penetrating radiation from the transmutation chamber 20, the active agent of which seems to consist in Neutrons. If the transmutation chamber is surrounded by a layer 21 composed : LA, Be, B, C, N, O, F, Ne, Na, Mg, Al, Si, of the elements P, S, Cl, A, K, Ca, Co, Ti, V, Gr, Mn, Fe, Co, Mi, Kr, Rb, Sr, Zr, X, Mb, Mo, Ru, Rh, Pd, Ar, Od, In, Sn, Sb, Te, I, Xe, Cs, Ba, La, Ce, Pr, Vd, Sm, Eu, 6d, Tb, Ds, Ho, Nr, Tu, In, Hr, Ta, W, Re, Os, Ir, Pt, Au, Hg, T1, Pb, B1, Th,U YD. H(2).

negative nuclei, for instance negative protons or diplots are generated in the layer. This is probably due to the action of the Neutrons of the mass 1, though part of the effect may be due to Neutrons of the mass 2 which may be generated by Neutrons of the mass 1 through their interaction with Diplogent. It can be determined for each element separately whether it generates negative nuclei, how much and of what mass and charge by means of a Wilson Choud Chamber, to which a magnetic field is applied. If we compose the layer 21 of an element "A", which generates negative nuclei, for instance negative protons, under the influence of the said penetrating radiation, and of an element "B", which transmutes into a radio-active element, under the influence of a negative nucleus radiation,

we can efficiently produce a new type of radio-active body

It is essential to have the bulk of element "A" so large that a large proportion of the Neutrons is utilised in nuclear collisions (layers of 10 c/m. to 100 c/m. will be needed) and it is further essential that elements "A" and "B" should be so mixed that many of the negative nuclei generated in "A" should reach "B" before or when they come to the end of their range. An example for a substance "A" which apparently yields negative nuclei, presumably negative protons, when bombarded by Neutrons, is Uran.

Some of the radio-active substances generated by negative proton bombardment are of an entirely new type; apparently the negative proton gets first captured in certain cases by the positive nucleus of the bombarded element without falling at once into the nucleus and subsequently there is a liberation of energy following an exponential law corresponding to the negative proton interacting with falling into the nucleus.

(Bombardment of hydrogen, diplogen, H(2), triplogen, H(3), by negative protons may lead to the generation of pseudo-neutrons, which may after a certain time framewite into real neutrons. Such bombardment may lead to the generation of pseudo-neutrons of the masses 2, 3 and 4 respectively, for which we shall use the symbols $\overline{n}(2)$, $\overline{n}(3)$ and $\overline{n}(4)$ to distinguish them from the real neutrons of similar mass numbers, for which we shall use the symbols n(3), n(3) and n(4). would

(Such pseudo-neutrons suffer energy losses through ionising matter (through which they travel) in a larger degree than is the case for real neutrons, and their range is therefore smaller. However, they can cause transmutation even if they have reached the end of their range and are therefore much more efficient in equating transmutations than protons, diplons or other positive nuclei. Negative nuclei have a range similar to positive nuclei, but they can cause transmutation even when at the end of their range and are therefore much more efficient in this respect than protons, diplons and other positive nuclei.

An important way of bringing about efficiently transmutation processes is to build up transmutation areas choosing the composition and the bulk of the material so as to make chain reactions efficient and possible, the links of the chain being "efficient particles".

One example is the following. The chain transmutation contains an element "C", and this element is so chosen that an efficient particle "x" when reacting with "C" may produce an efficient particle "y", and the efficient particle "y" when reacting with "C" may produce either an efficient particle "x" or another efficient particle which in its turn is directly or indirectly when reacting with "C" capable of producing "x". The bulk of the transmutation area, on the other hand, must be such that the linear dimensions of the area should sufficiently exceed the mean free path between two successive transmutations within the chain. For long chains composed of, say, 100 links the linear dimensions must be about ten times the mean free path. An example of such a chain reaction in which the efficient particles are neutrons and in which the element mis "C" is diplogen, any be the following :-

H(2) + n(1) = n(2) + H(1) + 4.5.M.E.V.H(2) + n(2) = H(3) + n(1)

In order to bring about such a chain reaction a large bulk of diplogen must be exposed to a neutron radiation generated as described above in Fig. I. The diplogen may be mixed with an element "B" chosen so that a neutron

In the above mentioned chain reaction an energy of about 4.5 M.E.V. is liberated while two atoms H(2) disappear and one atom H(3) and one atom H(1) are produced. We shall call a chain reaction at in which two efficient particles of different mass number or atomic number alternate a doublet chain. If both pasticles are neutrons we shall call it a neutron chain. An example for another doublet chain which is a neutron chain is the following reaction, which can be set up in a mixture of a neutron reducer reseiver element (like Lithium (6) or Boron (10) or preferably some of the heavy "receiver elements monthemed. in connection with "negative chains" on page /4), and "neutron a/converter element" which yields n(2) when bombarded by n(1). An example for such a chain in which carbon acts as Possiver and Beryllium acts as converter is the following:

C(12) + n(2) = C(13) + n(1)("Be(8)" need not mean an ex-isting element, it may break up spontaneously). "neutron Be(9) + n(1) = "Be(8)" + n(2)Diplogen ser act as a receiver as well as a/converter". If we have a chain reaction in which only one kind of efficient particle takes part we call the chain a singulat chain." We shall now give an example for a neutron singulet chain. Evidently A singulet chain can only be maintained in the presence of a metastable element i.e. an element which has sufficient energy to disintegrate STIM into parts the of the mass number of which is equal to its mass number or to transmute into an isomar under liberation of energy when meeting an efficient particle. An example for such a metastable element is Beryllium. When exposed to a neutron radiation Beryllium transmutes and increases the energy of the neutron with which it reacts:

Be(9) + n(1) = "Be(9)" + n(1) + Energy "Be(9)" would be if it $\frac{were}{15}$ stable an isomer of Beryllium

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but it need not be stable, it may disintegrate spontaneously into parts.

One can very much increase the efficiency of the hitherto mentioned neutron chain reactions by having a "neutro "O" multiplicator"/mixed with the elements which take part in *neutron*the chain reaction. A/multiplicator is an element which either splits up inke n(2) into n(1) + n(1) or an element for instance which yields additional neutrons/n(1) when bombarded by n(1). A multiplicator need not be thereastantly a metastable element but metastable elements and thereastable element but metastable elements and the berglium is the Berglium is the berglium is the thereastable element as multiplicators.

suitable multiplicator which yields neutrons under liberation

Be(9) + n(1) = "Be(8)" + n(1) + n(1) + Widdles:

Up till now we have only talked about pure neutron chains, we shall now talk about "mized chains' in which we have two efficient particles, one being a neutron, the or diplom other one being a negative proton forming the links of the chain:

 ${}^{"P"} + n(1) = {}^{"Q"} H(1)$ ${}^{2} {}^{"R"} + H(1) = {}^{"S"} + n(1) + Energy$

It is essential to chose the element "R" so as to second have energy liberation in the/process, whereas it is not necessary to have energy liberation in the first process. In order to be able to produce energy for power production and to store energy by producing radio-active bodies to be used as accumulators it is necessary to imme maintain a chain reaction (if the chain reaction is a neutron chain or a mixed chain) in a large bulk of material. This is due to the fact that the mean free path of the neutrons is large. The situation is entirely different for pure negative chains with which we deal further below.

A mixed chain in which n(1), n(2) and N(1) take part (a triplet reaction) could be maintained in a mixture of H(2), H(1) and L2(7) L1(7) + n(2) = Ho(4) + Ho(4) + H(1) + Energy H(1) + H(1) = n(2) size L1(7) and other receiver elements contioned further below iny yield negative protons when bombarded by n(2). Figure shows an example for utilizing neutron

chains and mixed chains for power production and the generation of radio-active bodies. 101 is a high voltage positive ray tube generating fast light ions like diplons or Helium ions which cause by striking diplogen or Beryllium in 102 the emission of a penetrating radiation (neutrons). The radiation emerging from 102 acts on the material 103 which forms a sphere around 102. This material is such that a chain reaction, preferably accompanied by the action of a multiplicator is released. For instance we can have a sphere of Beryllinne 105, the dimensions of which are so chosen that the energy liberated in the Beryllium should be a multiple of the energy input. The pumps 120, 121 and 122 pump a liquid for instance water or mercury through the 107, 110, 111 pipe systems IXEX 102, 197, 199 andxiderapipersystems 103 thereby cooling the transmutation area and driving the heated liquid through the boiler 126. The boiler supplies annual steam to a power plant. The neutrons emerging from the sphere 103 act on a layer 104 which is composed of an element/that will transmute into a radio-active body which is suitable for the storage of energy. > The element "T" need not be present as a free element but can preferably be present in the form of a compound solupple in water; active that makes it easier to separate the radio- bodies formed in the process. A third layer 105 contains an element "V" that will absorbe the neutrons /n(1)/ under liberation of energy (Li(7) or some other "receiver" element). 106 is a heat insulating layer.

Negative Chains.

9.

This paragraph deals with chain reactions in which negative protons or negative diplons or both are engaged.

Negative protons can be generated by shooting neutrons on one of the elements mentioned on page 3. In order to determine which element or which isotope of a given element is most efficient in yielding negative protons (or negative diplons) when bombarded by neutrons, one has to investigate each element or each isotope respectively by means of a Wilson Cloud Chamber to which a magnetic field is applied. Instead of the Wilson Choud Chamber one can also use an ionigation chamber in connection with an amplifyer and an oscillograph and use a magnetic field to deflect the "protons" before they enter the chamber.

If one exposes a mixture of Diplogen and another element which acts as a "reducer" such as for instance Lithium (6) or Reryttinx Boron (10) or Carbon (12) to the action of negative protons or diplons, chain reactions of the following type may be maintained: LI(6) + H(2) = Li(7) + H(1) + EnergyH(2) + H(1) = H(1) + H(2)

As we see energies liberated in the first reaction, but there is no liberation of energy in the second reaction. The latter fact makes this second reaction objectionable for the following reason: if we have a pure neutron chain which is a doublet chain reducer it is not important that both the **RECEIVER** and the **REX** converter **ARGEN** MERLEN reaction should yield energy, but if we use a pure negative chain which is a doublet chain it is reducer important that both the **RECEIVER** and the converter reaction should yield energy. d. Diplogen itself can act as a " volucer"

The above mentioned reduces had therefore better be used in combination with another " converter" than diploger. proton rediction.

In order to get a multiplicating action a third element "E" can be mixed to the two element mixture which maintains the chain, the element "I" may chow the following reaction

z E'(m) + H'(2) = F'(m) + H(1) + H(1) + ln. In order to determine which element is capable of this reaction a source which yields negative diplons can be surrounded subsequently by thin sheets of the elements mentioned on page 3 and by means of a Wilson Cloud Chamber or an iodisation chamber of the type, in combination with the magnetic field one can determine which of the said elements yields sufficiently large number of negative protons when bombarded by negative diplons. May element 'E' will be called an under hay ed multiplicator.

We shall call subsequently two elements which have the same mass number fisobar pairs of the first order if their atomic number differs by one. Such isobar pairs are for instance

In(115)-Sn(115; Stores Perios); An(197), Hg/10 Te(205) Pb(103); Pl(205) Pb(205); The mass of the two elements forming an isober pair is

not exactly equal. In certain cases the mass of the element of the lover atomic number is larger than the mass of the sus element with the higher atomic number. If the mass difference is sufficiently large a negative diplon striking the element with the lower stomic number can get also in the case split up into two negative protons initial kinetic energy of the negative diplon parto wee negligible). In this way an element which forme, is " undercharged isobar pair of the first order and nts forming the pair may be a suitable sultiplicator in a chain reaction in which gin such cases we shall dall the element of the lower stomic number en undercharged element of the first order."

negative diplons and negative protons alternate. An underemarge: element of the first order will be called an undercharged multiplicator if its mass is sufficiently high in respect to its partner forming the pair to split up a negative diplon into two negative protons in accordance with the law of the conservation negative of energy if the/diplon is at rest. (The mass difference between the negative diplon and two negative pro-0015 for 0.602 tons being .005, the mass difference between the two elements forming the isobar pair of the first order must be at least as much).

If we use the word "undercharged element" necessarly processor we do not necessarily mean an element which belongs to all so an isobar pair (doublet) but he meen any element which can t rensmute under liberation of energy while its charge (atomic number) increases (or more precisely while the sum of the charges of the parts into which it may fall in the process increases). We shall talk of undercharged elements of the first, second or any higher order accordingly whether energy is liberated while the charge increases by it one, two or more units. Evidently an undercharged element can be both of the first and second order etc. An example of an undercharged element is for instance potassion (E). the radivaletive isatope of putto (K)

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As we call all elements metastable which kave a antitiziently large mass have a sufficient energy to transmute under liberation of energy while the mass number does not change (therefield of the mass numbers misthe parts into which it may form does not change). and the sum of the mass numbers of the parts is equal to the original mass number.

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uccosserily consider all undercharged elements Examples for metastable elements are motestable. the radio-active isotope of further/potassium (X), Uran, Thorium and Beryllium.

A(40), Ca(40); esc. and/the mass of the element with the smaller atomic muber is larger than the mass of its partner in the doublet, the former one is necessarily an undercharged particle of the second order. doublet of the pourth order

If we have a isobar triplet like for inst

and if the mass of the element which has the lowest abomic number is smaller than the mass of the element which has the highest stanic number in the tr the former one is necessarily an undercharged element of the finth order.

We shall now some back to discuss the chunge chain reactions in which negative diplons and negative protons alternate. We have dealt with such reactions in mixtures of diplogen and some other element (Li(6), B(10), N(14) in which the partner of the diplogen has been selected among those elements of which we elreedy know that they yield fast protons when bonberded by fast diploms (positive diplons) under liberation of energy. We state call such elements "radman" anda it is say the first must which yield under liberation of energy negative protons when bombarded by negative divions "roducer" elements, and it is easy to find out by experiment which of the elements mentioned on page There are only few 20 10 pope allesionite is the most officient reducer.

to find elements which yield negative diplons when bombarded by negative protons without swallowing much energy in the process. Such elements will be called "converters" elements, and in the chain reaction described further above diplogen played the role of the converter element. Diplogen does not swallow energy in the process of convertion, but neither does it liberate energy in the process. A converter element which liberates energy in the process of yielding negative diplons when bombarded by negative protons, will be called an "abundant converter". Such an abundant converter is for instance Beryllium.

It is an advantage to select the mixing two elements, a "reducer" and an "abundant converter" to form the mixture in which a chain reaction is produced, possibly in the presence of an undercharged multiplicator (element "E").

We can use for instance Lithium (6), Boron (10), Hitrogen (14) etc. as "reducer" in combination with Beryllium (instead of diplogen) as Cabundant converter". <u>According to which of the abovementioned elements</u> we use in connection with Beryllium we get in suffice reactions of the following type: L1(6) + H(2) = L1(7) + H(1); Be(9) + H(1) = Be(8) + H(2) + En. B(10)+H(2) = B(11) + H(1) En.; Be(9) + H(1) = Be(8) + H(2) + En. N(14)+H(2) = N(15)+H(1) En.; Be(9)+H(1) = Be(8) + H(2) + En. Q(14)+H(2) = C(13)+H(1) En.; Be(9)+H(1) = Be(8) + H(2) + En.

of course Be(8) need not be stable but can fall at once into parts of transmite after some time. The above mentioned reaction between Be(9) and H(1) can also be written in another form in order to indicate that the negative proton may be captured by the Beryllium nucleus and the newly formed nucleus may disintigrate subsequently with a certain half period. In order to indicate this possibility we canutite the reaction in the following form

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 $Be(9) + H(1) = "_3L1(10)"$ "_11(10)" = "_Be(8) + H(2) + En

In order to get a chain reaction in which $\tilde{H}(2)$ and $\tilde{H}(1)$ alternate we have to use as montioned above a converter and a roducer. Apart from the reducers mentioned above suitable reducers can be found among the elements which have an even a tomic number and have at the same time an atomic weight \geq close to a whole number. The latter fact indicates that the element is mainly composed of one isotope. Such elements are O16) Ne(20), Ng(24), S1(28), S(32), Cr(52), A(40), 71(48), Ba(138), Ge(140), Th(232) U(230). Especially suitable are of these elements those which have a weak isotope of an atomic number which exceeds by one the atomic number of the main isotope; such elements are 0, Ne, Ng, S1, S, and Cr.

We shall subsequently call chain reactions in which two efficient particles alternate (for instance two negative hydrogen isotopes) doublet chains. As a rule a mixture of at least two elements is needed to maintim a doublet chain unless one and the same element acts as converter as well as reducer. It is preferable to use a metastable element as converter, and essential to use a metastable element as multiplicator (if we wish to have a multiplicator acting in the process), if we want to split negative up a hydrogen isotope. This is essential for the following

reason: the negative hydrogen isotope for instance/11(2) requires energy in order to be able to split up in two negative protons. This energy may be supplied by the kinetic energy of H(2) itself, but in view of the fact that the range of H(2) is small only wary for particles can be anith up a small fraction of the H(3) perticles can split up in this way before the end of the range Once theH(2) particle loges its kinetic is reached. meeting a motastable energy it can only be split up when element "E". In order to indicate that it may first get captured by that element and solit up subsequently with a certain half period we simil/write the reaction (which has already been mentioned above) in the following form:

 ${}^{"}_{2}E(n)^{"} + H(2) = {}^{"}_{2} D(n 2)^{"}$ ${}^{"}_{2} D(n 2)^{"} = {}^{"}_{2} F(n)^{"} + H(2) + H(2)$

In these equations z means the abomic number and m means the mass number of the undercharged multiplicator "E". Beryllium can act both as a convertee and an undercharged multiplicator.

There are not many metastable elements among the light elements which can act as converters or multiplicators or both, but such elements may be found among the metals which have a mass number above 120 and an atomic number above 50. Of such elements those are the most suitable which have an atomic weight close to a whole number, a fact which indicates that the element contains mainly one isotope. Such elements are Uranium (U) Thorium (Th), which are known to be metastable, further iridium (Ir). Tantal (Ta), Bismuth (Bi), Gold (Au), Ce, Ba, Cs.

In a mixture of a receiver and a metastable element in which we wish to maintain a doublet chain the metastable element will have to be present in abundance, i.e. the mixture will contain comparatively little of the receiver element.

We shall now say a few words about singulet chains which can be induced in metabtable elements, for instance in Beryllium if negative protons or diplons are acting on Beryllium. These are chains in which energies in liberated by an efficient particle which does not disappear in the process, nor does it change its mass or mass charge (minute/number or atomic number). Such a chain can be maintained in Beryllium, the links of the chain being negative protons:

 $_{4}^{4}Be(9) + H(1) = _{3}^{4}be(10)^{*}$ $_{3}^{4}be(10)^{*} = _{4}^{*}Be(9)^{*} + H(1) + Energy.$ We see that the negative proton which has been captured by Beryllium nucleus is released under liberation of energy while the Beryllium transmites into "Be" (which in its turn may break up spontaneously) and the released negative proton is free to react with another Beryllium muleus.

A similar singulet chain can be maintained in Beryllium using negative diplons instead of negative protons.

Other metastable elements may take the role of Beryllium in such singulet chains, in which either negative protons or one of the negative hydrogen isotopes form the links of the chains

" $_{2}$ M(m)"4 H(2) = $_{7-2}^{n}$ L(m+2)" "L(m 2)" = " $_{7}$ K(m)"+H(2) + Energy In this equation m means the mass number of the metastable element "M" which transmites under liberation of energy into an isomer "K" (which may or may not disintigrate

sponteneously in its turn).

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The following are essential features of the production of power, the storage of ower by means of radio-active bodies, and the production of radioactive bodies in general by means of chan reactions in which the links of the chain are formed by negative nuclei for instance negative protons or negative diplons or both.

1. Shoulet chains. The exposure of a metastable (or element to a negative proton, negative diplon) source or to a neutron source, the radiation of which will generate either in the metastable element itself or in element "A" mixed with the metastable element, negative protons or other negative nuclei.) For instance the exposure of a Boryllium layer to a negative proton source. Means to cool the layer of the metastable element and means to utilise the heat obtained for power production.

2. Doublet chains. The exposure of a substance to negative proton or negative diplon action as in case of point 1. but the substance being chosen so that it should be a receiver and a converter, for instance a mixture of two elements, one of which is a receiver, the other a converter. Means to utilise the heat obtained like in the case of singulat chains. for instance Beryllium as The use of a metastable element as/converter. Chains with miltiplicating action. 3. The exposure of a substance to a negative protons (or other negative nuclei for instance negative diplons) the substance being so chosen thes it should act as an undercharged which acts miltiplicator. The use of a substance bath as a multiplicator as well as a converter and a for instance the use of a mixture of three receiver: elements each of which has one of the mentioned actions. Heans for usaveying and utilizing the heat liberated in the layer in which the chain reaction takes place (the transmitation layer). The exposure of an animatumes element "B" which transmites into a radio-active body under the action of negative nuclei generated in the transmitation layer to these nuclei by surrounding the transmitation layer with such an element or mixing such an element to the transmitation layer.

Figure 4 shows on example of inducing and utilising such chain reactions. 201 is a layor forming a hollow sphere, the transmitation layer in which the chain reaction takes place. We have in the middle of the sphere a source/of negative prokins or tha/source 202 of a penetrating radiation for instance neutrons which will produce negative protons when penetrating the transmitation layer 201 which may contain an element "A" that yields negative protons when bombarded by neutrons. The sphere within the layer 201 is not only hollow but a reduced gas pressure (vacuum) is maintained in it; so that negative nuclei emitted by the layer 201 towards the interior of the sphere should strike the layer again at some otherpoint of the inner surface. The outer surface of the layer 201/is siths formed a drawn in the upper right quadrant of the cross section shown in Firre 4 . This has the purpose to stabilise the chain reaction if we have to deal with the chain reaction in which multiplicating action is involved, so as to be able to reach a high multiplicating factor without danger of Layor 203 absorbes the negative nuclei explosion. emerging from layer 202 and can be formed by substance "B" which will transmite into a radio-active body, thereby storing energy. The left upper quadrant shows enother form of the outer surface of layer 201 which serves the

same purpose as the form shown in the upper right quakrant. 204 is a high voltage positive ray tube which is used to accelerate diplogen or helium ions which produce neutrons when they hit the target 205 consisting of diplogen or beryllium. These neutrons liberate negative protons from the layer 206 (substance "A") which reach through the vacuum the inner surface of layer 201. If layer 201 contains a substance "A" further negative protons may be liberated in the layer 201 itself.

It is also possible to use negative protons or negative diplons an negative fixing by shooting positive protons or positive diplons on undercharged It is alloged that elements of the second order. the radio-active icotope of potassium is an undercharged element of the second order. It is also possible to produce negative protons or diplons by shooting positive triplogen, or halium ions or one of the elements mentioned on page 3 which need not be to should an undercharged element for inspance Lift an O(18) www.weinerenthink An efficient populcie therefore disappears (and a chain is/interrupted if this happens in a chain reaction) when a negative proton of a neutron reacts with a namein nucleus in such a way that the regative proton or the neutron disappears and a positive particle for instance a proton of en alpha particle ere If we wish to produce negative protons or ather emitted. negative by neutron sombardment of cortein elements we can surpress such an occurate by bombarding heavy Slow neutrons can be elements with slow neutrons. produced for this purpose by disintigrating diplogen (by shooting diplogen ions on it or by heating up the procise the can surpress the dislogen). production of a positive particle when bombarding the elemen

by neutrons by choosing the element and the neutron the creation of which has energy so that the positive particle/which has a potential possibility should not have sufficient energy at its disposal to penetrate in the inverse process the nucleus of that element. The same holds for the discoverance of negative protons accompanied by the creation of a positive particle. In order to avoid such an occurance in our chain reactions we shall use as reducers, converters and metastable multiplicators or notastable singulat chain maintaines the heaviest elements which are otherwise satisfactory. Form P. Ack. 4



THE PATENT OFFICE,

No. 19157 1,28 JUN. 1934

25, Southampton Buildings, Chancery Lane, London, W.C.2.

Received documents purporting to be the Application and

Provisional Specification of

which have been numbered and dated as above,

M. F. LINDLEY,

relard

Comptroller-General.

N.B.—Unless a Complete Specification is left on an Application for a Patent within <u>TWELVE MONTHS</u> from the date of application (or with extension fee, 13 months), the Application is deemed to be abandoned. The investigation as to novelty prescribed by the Patents Acts, 1907 and 1932, is made only when a Complete Specification has been left.

The number and date of this Application must be quoted on the Complete Specification and Drawings (if any), as well as in any correspondence relative thereto.

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Patents Form No.3.

Provisional Specifications

19157/34 19721/34 27507/34	Dated N	28th 4th 25th	June July Sept.	1934 "
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PATENTS AND DESIGNS ACTS, 1907 to 1932.

COMPLETE SPECIFICATION

Improvements in or Relating to the Transmutation of Chemical Elements.

I LEO SZILARD, a citizen of Germany, a subject of Hungary, c/o Claremont Haynes & Co., of Vernon House, Bloomsbury Square, London, W.C.l. do hereby declare the nature of this invention and in what manner the same is to be performed, to be particularly described and ascertained in and by the following statements:-

The invention concerns methods and apparatus for the production of nuclear transmutations leading to the generation of radio-active bodies, to the liberation of nuclear energy and the untilisation of the liberated energy.

Stamp. £4. 9.4.35. According to this invention, neutrons of a higher mass number (I have reason to believe that such multiple neutrons which carry no charge and have a mass roughly equal to a multiple of the proton mass, exist) are generated in a chain take part reaction in which neutrons of different mass number/and energy or radio-active bodies or both, are generated either in the process of the chain reaction itself or by the radiations arising out of the chain reaction.

There are several radiations arising out of chain reaction which may generate radio-active bodies, for instance, radiation consisting of neutrons of mass number 1; radiation consisting of neutrons of mass number higher than 1 (multiple neutrons) and gamma radiation. I wish to make it clear that methods and apparatus for the generation of radio-active bodies by means of neutrons of mass number 1, without chain reactions, in itself is not claimed, and does not form part of the subject matter of this specification. It forms part of the specification and is claimed in my application of letters patent No. 7840/1934.

In the chain reactions to be described below, energy is liberated in the form of heat and can be utilized for power production by making use of the heat liberated in the chain reaction. Through the generation of radio-active bodies energy is being stored and gradually liberated in the form radiations which can easily be transformed into heat, which can be utilised for power production. Furthermore, the energy stored in the form of radio-active bodies can also be more directly utilised for the generation of electricity since radio-active bodies emit electrically charged particles and thereby may directly generate electrical energy.

In the following I shall deal with methods and apparatus for the production of energy and the generation of radio-active

bodies by means of chain reactions. In order to maintain such a chain an initial radiation of neutrons my be generated, for instance by one of the methods described in may Specification and Application for letters Patent No. 7840/1934. If the neutrons enter a space which has the proper shape and size and is filled with the proper combination of elements the energy or the number of the neutrons, or both, can be greatly increased through their interaction with the substance which fills the chain reaction space. The interaction of a neutron with matter can lead to the liberation of a multiple neutron this multiple neutron liberates in its turn one or more neutrons of mass number 1 which in their turn liberate again multiple neutrons. In this way we can maintain a chain reaction in which a large number of neutrons and multiple neutrons are liberated, the total number being determined by the geometry of the arrangement.

Figs. 1 and 2 show such a chain reaction apparatus. A neutron radiation, the initial radiation, is generated by the high voltage canal ray tube 1, Fig 1. 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, Fig 2 pump a liquid for instance water or mercury through the pipe systems 107, 110, 111, Figs 1 and 2 thereby cooling the transmutation area 3, Fig 1, and driving the heated liquid through the boiler 126 Fig 2. The boiler may supply steam to a power plant. The neutrons emerging from the sphere 3 act on a layer 9, Fig. 1 which is composed of an element that will transmute into a radio-active body.

An essentially different way of introducing the initial radiation into the chain reaction chamber is the arrangement shown in Fig 3. 1 is the cathode ray tube 402 is a sheet of heavy element for instance Pb, or U in which penetrating radiation hard (X-rays) is generated with good efficiency if the electrons have a voltage of over one million volts. This efficiency increases very rapidly with the voltage, and is much higher than 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 consisting of the layers 407 and 3 (for the cooling of this chamber and the utilisation of the heat generated in it I refer to Fig. 2, 106 in Fig 3 is to be identified with 106 in Fig. 2.) Nevertheless the sheet can be sufficiently thick to utilize more than half 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 beryllium is present in 407 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.

I shall demonstrate in the following the importance of the shape and the size of the transmutation space. I assume that the chain reaction takes place in a closed spherical layer of material the inner radius (r.) of which is large compared with the mean free path (a) of the neutrons (or other particles which are involved in maintaining the chain). In the simplest

case the density (s) of the neutrons will with good approximation be given as a function of the radius (r) by the following equation:

D. d(rs)/dr + A. (rs) = 0

D and A are determined by: the mean free path (a) of the neutrons; the mean velocity of the neutrons w; the factor of the multiplicating action f which says how many collisions of a neutron are needed in the average in order to produce one new neutron.

A = w/af; D = aw/3; $\sqrt{\frac{D}{A}} = \frac{a\sqrt{f}}{\sqrt{3}}$

I am interested in the critical thickness of the spherical layer for which the gradient of the density s vanishes for the internal radius (r.). If the thickness of the spherical layer $(r_2 - r_1)$ $(r_2$ and r_1 are the external and internal radii respectively) approaches a certain critical thickness L one can maintain with a very weak source of initial radiation in the interior of the inner surface of the spherical layer a very strong chain reaction and one can easily get one thousand or more times more neutrons emerging from the Chain reaction layer than the number of the neutrons forming the initial radiation. If the outer surface $(r = r_2)$ of the spherical layer were to stand free in space the density s would be zero for that surface and the critical value L would be given by L = $\frac{\pi}{2} \sqrt{p/A}$ If the outer surface is covered by some material, for instance if the transmutation layer is immersed into water or covered by lead the critical value L is This is due to the back scattering by water or lead reduced. and also to the fact that the neutrons are slowed down in the water and their mean free path is thereby reduced.

It is important to prevent neutrons from escaping out of the interior of the inner surface of the spherical layer and also from being absorbed in the interior. If the initial radiation is generated by apparatus placed in the interior of the sphere the material used should be so selected as to lead to a minimum of absorption.

If the thickness of the layer is larger than the critical thickness L the number of neutrons would go on increasing indefinitely and such an increase is only stopped when the heat which is liberated in the process causes the spherical layer to explode.

The differential equation which I have given above and from which we have derived the value for the critical thickness L does not give the correct description of the density of the neutrons in a chain reaction, nor does it give the correct In order to get the correct equation we have value for L. obviously to distinguish between the mean free path a, of the neutron for a collision and its factor f, which says how many collisions of a neutron are needed in the average in order to produce a multiple neutron on the one hand, and on the other hand between the mean free path ag of the multiple neutron and its factors fo and fo of its multiplying action which says how many collisions on the average of the multiple neutron are needed in order to produce one or two new neutrons respectively The only purpose of putting down the above simplified etc. equations was to demonstrate the general type of behaviour of chain reactions with multiplying action and to sow the existence of a critical thickness L. The simplified equation is an approximation of the correct equation if many collisions of the neutron are needed to generate a multiple neutron but few collisions of the multiple neutron are needed to generate two neutrons

I shall now discuss the composition of the matter in which the chain reaction is to be maintained. It is essential that two different heavy non-positive particles should take

part in the reaction in order to obtain the chain. (Heavy non-positive particles are particles which have a mass roughly equal to the mass of the proton or a multiple thereof and which carry no positive charge. The neutron is such a particle, its mass being roughly equal to the mass of the proton, and its charge being 0. I have reason to believe that heavier isotopes of the neutron exist, the mass of which is a multiple of the proton mass and the charge of which is 0.)

A mixture of two elements "E" and "P" can be so chosen that element "F" (the converter element) when it reacts with a simple neutron should transmute into an element the mass number of which is lower and generate a multiple neutron; on the other hand element "E" (the reducer element) should when it reacts with a multiple neutron transmute into an element the mass number of which is increased and generate a simple neutron. In order to have a chain reaction in which the number of neutrons increases it is necessary that apart from the converter and the reducer element there should be present a multiplicator element that is to say one from which neutrons are liberated by neutrons in a process in which the interacting neutron is not captured or alternatively a multiplicator element which generates four neutrons from a multiple neutron.

I wish to give the following indication of which elements may be used as converter "F": The fact that an element ejects a multiple neutron, for instance a tetra neutron (a neutron of mass number 4), when bombarded by simple neutrons, can be revealed by the fact that it becomes radio-active through neutron bombardment, and that the generated radio-active element is an isotope of the bombarded element itself. For instance, if indium is bombarded by fast neutrons (of less than 8 M.¹E.¹V. energy, but more than 100,000 E.V. energy) a radioactive isotope of indium is generated, which decays with a 4 $\frac{1}{2}$ h.

period. This indicates that one stable indium isotope captures a neutron, and a multiple neutron is ejected, leading to a radio-active indium isotope of mass number 112. Probably the stable indium isotope 115 captures the neutron and ejects a tetra neutron. Another example is bromine from which very slow neutrons generate three radio-active bromine isotopes, two of these can be accounted for by radiative capture of the neutron, but one of them probably arises from the stable bromine isotope of mass number 81 which captures a simple neutron, ejects a tetra-neutron leading to a radio active isotope of bromine of mass number 78. Only very few elements will eject a tetra neutron when bombarded by very slow The number of elements which can eject a tetra neutrons. neutron increases with the kinetic energy of the bombarding Not all the elements reveal this fact by an simple neutron. appreciable radio-activity, therefore a more general method can be employed to investigate each element separately. This more general method is based on the detection of the ejected tetra neutron. The ejected tetra neutron can be detected through the transmutation which it causes in various elements which are exposed to it. Such transmutations reveal their presence in two different ways; either through radioactivity induced in the element which is exposed to the tetra neutron, or through the ejection of charged particles (proton or alpha-particle etc.), from the element which is exposed to the tetra neutron. The ejection of such charged particles can be observed by means of an ionization chamber, a Wilson cloud chamber or a photographic plate which contains the element, which transmutes when exposed to the tetra neutron.

I further wish to give some indication as to which elements may be used as reducer element "E", from which a multiple neutron liberates a simple neutron, and as multi-

plicator element, from which a multiple neutron liberates two simple neutrons.

A lower limit for the mass of the tetra neutrons can be deduced from considering two radio-active elements, of which the lighter one arises from the heavier one, through two beta transformations and one alpha transformation. If the mass of the tetra neutron were smaller than the mass differences of these two radio-active elements, the heavier elements would spontaneously have to eject the tetra neutron, and would thus spontaneously transmute into the lighter element.

By applying this consideration to the known radio-active elements, we obtain as a lower limit for the mass of the tetra neutron about 4.014. While the slow neutron will eject a tetra neutron from only few elements, a tetra neutron having such a high mass will eject a neutron from most of the elements and will eject two neutrons from a number of elements. In order to determine from which elements it ejects two neutrons (multiplicator elements) we have to take each element in its turn, bombard it with tetra neutrons and either observe the number of simple neutrons which emerge, or observe the radioactivity induced in the bombarded element, and thereby identify the nature of this transmutation. An example for a multiplicator element of this type might be beryllium, and many elements heavier than beryllium. Heavy multiplicator elements are as a rule preferable since they will emit no, or few, positively charged particles, and we can thereby avoid interruptions of the chain.

The value of the critical thickness "L" previously referred to, can be estimated for a spherically symmetrical body as follows: The mean free path for an elastic collision of the neutron is in many elements of the order of 5 cms. Every hundredth elastic collision may lead to the

ejection of a tetra neutron, and every collision of the tetra neutron (mean free path of the order of 5 cms) may lead to the ejection of two simple neutrons. In these circumstances "L" will be of the order of magnitude of 50 cm.

By maintaining chain reaction in combination with means away for leading/and utilizing the heat set free in the transmutation process energy can be produced and utilized for power production.

Having now particularly described and ascertained the nature of my said invention and in what manner the same is to be performed, I declare that what I claim is :-

- 1. A method for the generation of uncharged particles the mass of which is roughly equal to the proton mass or a multiple thereof, which generated neutron isotopes generate radio-active elements or energy or both, characterised by the maintenance of a chain reaction in a body in which neutron isotopes of different mass number take part.
- 2. A method according to Claim 1 characterised by a chain reaction in which a neutron of mass number 1 and a heavier neutron isotope take part.
- 3. A method according to Claim 1 or 2. characterised by the generation of an initial radiation which can consist of neutrons of mass number 1, and the exposure to this initial radiation of a body so composed that a chain reaction is caused by the initial radiation.
- 4. A method according to Claims 1, 2 or 3 characterised by the said body containing a converter element and a reducer element.
- 5. A method according to Claims 1, 2 or 3 characterised by the said body containing a converter element and a multiplicator element.

- 6. A method according to Claims 1 2 or 3 characterised by the said body containing a converter, a reducer and a multiplicator element.
- 7. A method according to Claims 1 2 or 3 characterised by the said body containing beryllium.
- 8. A method according to Claim 1 2 or 3 characterised by the exposure of an element to the radiations generated in the said body which element transmutes into a radioactive element under the influence of the radiations generated by the chain reaction.
- 9. A method according to Claims 1 2 or 3 characterised by the use of a hydrogen containing substance, for instance water, for scattering the neutrons, for instance using water and surrounding the whole body in which the transmutation takes place by water.
- 10. Improvements in or relating to the transmutation of chemical elements by means of a chain reaction as hereinbefore described and illustrated in the accompanying drawings.
- 11. An apparatus for carrying out the methods claimed in any of the Claims 1 to 9 as hereinbefore described in the specification and shown in the accompanying drawings, or any other apparatus which is substantially equal thereto.

Dated the 9th day of April 1935.

CLAREMONT HAYNES & CO.

Vernon House, Sicilian Avenue, Bloomsbury Square, W.C.

Applicants Solicitors.

IMPROVEMENTS IN OR RELATING TO THE TRANSMUTATION OF CHEMICAL ELEMENTS.

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DR. LEO SZILARD

COMPLETE SPECIFICATION.



28 Jun. (Fig 2 of original 7840 ?) 1 Shift and



Fig. 4.

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few, positively charged particles, and we can thereby avoid interruptions of the chain.

Other examples for elements from which neutrons can liberate multiple neutrons are uranium and bromine .

The value of the critical thickness "L" previously referred to, can be estimated for a spherically symmetrical body as follows: The mean free path for an elastic collision of the neutron is in many elements: of the order of 5 cms. Every hundredth elastic collision may lead to the ejection of a tetra neutron, and every collision of the tetra neutron (mean free path of the order of 5 cms) may lead to the ejection of two simple neutrons. In these circumstances "L" will be of the order of magnitude of 50 cm.

By maintaining chain reaction in combination with means for leading away and utilizing the heat set free in the transmutation process energy can be produced and utilized for power production.