

According to this invention, a nuclear chain reaction can be maintained in a system which contains a heavy element that undergoes fission under the action of slow neutrons, (such as, for instance, natural uranium) and a light element of a class defined further below, which slows down the fast neutrons emitted during the process of fission by the said heavy element.

*The slowing agent containing the light element*

Natural uranium is an example for a heavy element that undergoes fission with neutrons that have been slowed down to thermal energies. Fission induced by slow neutrons in natural uranium is attributed to the rare isotope U235, contained in natural uranium. Fast neutrons, such as are emitted in the fission of U235, cause fission also in the abundant isotope, U238, which is contained in natural uranium.

Carbon, particularly in its pure form of low ash graphite, is an example for a light element that can be used for slowing down the neutrons in a potentially chain reacting system. Let us consider an infinitely extended system <sup>of suitable</sup> at a given composition and structure. On the average of the neutrons emitted in a fission process, a fraction  $q$  is absorbed within the system as <sup>a</sup> slow neutrons <sup>(thermal or quasi-thermal)</sup> in such a manner as to produce fission, and on the average  $\eta$  fast neutrons are emitted for every fission process <sup>such such</sup> induced by slow neutrons. The system is potentially chain reacting if the product  $q\eta$  is larger than 1.

In order to have an actually chain reacting mass, we must build a sufficiently extended system. If the system is not surrounded with much material which efficiently reflects neutrons, the neutrons can escape from the surface of the chain reacting mass, and in that case, the critical radius  $l$  for which a spherically shaped mass becomes chain reacting, is given by the formula

(1)

$$l = \pi \sqrt{\frac{F^2}{3(\mu g - 1)}} \sim \sqrt{\frac{3F^2}{\mu g - 1}}$$

*overabundant  
have a density  
of at least  
0.5 gm/cm<sup>3</sup>  
in order  
to keep the  
kinematics  
within  
practical  
limits.*

①

*other elements*

*one or in form of a compound  
may be solid or liquid*

*related to -2-*

In this formula,  $\bar{r}^2$ , is the average square of the distance to which a fast neutron emitted in a fission process diffuses within the system until it gets slowed down and ~~causes fission~~, *is absorbed, and has for graphite of 1.7 gm/cm<sup>3</sup> of density a value of (50 cm)*

For a system the composition which will be described below, and which contains a lattice of aggregates of uranium embedded in graphite of a density of about 1.6 gm/cm<sup>3</sup>, a sphere containing about 500 tons of *pure* graphite is sufficiently large to maintain a chain reaction. *so that we have* If in place of a spherical shape, a cylindrical shape is used, and if the diameter of the cylinder is *R ≈ √(7500 / πg-1)* about equal to its height, the critical mass is about 10% higher.

For any given shape the critical mass is defined as the mass at which under the given condition, the intensity of the chain reaction just remains stationery. If the critical mass is exceeded, the intensity of the chain reaction (which manifests itself in the intensity of the neutron *and* gamma radiation emitted from the chain reacting pile), increases exponentially with time. If the mass is lower than the critical mass, but close to it, and if we have a source of neutrons (initial neutron radiation) within the mass, the number of neutrons produced in the chain reaction can be made larger than the *number of the* neutrons of the initial radiation as the mass approaches *more and more steeply* by a factor which increases to infinity to the critical mass. In order to maintain a chain reaction, it is not necessary, however, to make use of an artificial source of neutrons, since initial neutrons are always present; they are for instance emitted in the spontaneous fission of uranium contained in the chain reacting mass. *and present in the concrete caps*

The critical mass can be varied by introducing a slow neutron absorbers, *nickel or cobalt/nickel* such as cadmium or a cobalt iron alloy, into the interior of the mass, or by withdrawing the absorbers therefrom, by which means the critical mass is increased or decreased, respectively. This can be used for controlling *the* chain reaction, for instance, by moving the slow neutron absorber in and out in such a manner as to *have* be part of the time below, and part of the time above, the critical conditions. *the system*

In order to determine empirically the critical mass such slow neutron absorbers may be built into the system while the system is being <sup>assembled</sup> built up, and after a sufficiently large mass is assembled, these absorbers may be one after the other withdrawn. While these absorbers are being withdrawn, the radiation emanating from the mass may be observed, and ~~their~~ <sup>these</sup> position <sup>of the absorbers may be</sup> noted at which the neutron intensity begins to rise exponentially with time, which signifies that the critical condition has been reached.

*Of the neutrons emitted in fission*  
 A fraction  $\eta'/\eta$  which is close to 1 is emitted instantaneously and only a small fraction  $1-\eta'/\eta$  is emitted with a time delay of a few seconds. Though this delayed neutron emission is small, it has an important effect on the rate at which the neutron intensity rises exponentially with time if the mass of the chain reacting unit ~~slightly~~ <sup>only slightly</sup> exceeds the critical mass. In order to discuss this we may introduce the concept of the "critical mass for instantaneous neutron emission" which corresponds to the value of  $\eta'$  while the critical mass for the total neutron emission corresponds to  $\eta$ . This <sup>e</sup> critical mass for instantaneous neutron emission is larger than the critical mass. We can avoid a rapid exponential rise of the chain reaction intensity by keeping conditions so that while we may exceed the critical mass, ~~we~~ <sup>the system</sup> should remain in the interval between the critical mass and the critical mass for instantaneous <sup>neutron emission</sup> fission. Preferably in order fully to profit from the effect of the delayed neutron emission, ~~we~~ <sup>the system</sup> should remain within the interval between the two critical masses close to the critical mass for the total neutron emission.

.... *stop!!*

As will be stated further below, a chain reaction can be maintained by means of ordinary uranium in a system in which carbon in the pure form of a low ash graphite is used for slowing down the neutrons. Carbon has an absorption

but we can still leave a chain reaction in a system of otherwise favorable composition and structure.

cross section for thermal neutron of about  $\sigma_c = 5 \cdot 10^{-27} \text{ cm}^2$ . If impurities are present, the absorption cross section of the slowing agent per carbon atom is larger and we may allow it to rise perhaps as high as about  $10^{-26} \text{ cm}^2$  before it becomes impossible to maintain a chain reaction in a system of otherwise favorable composition and structure.

Carbon and other light elements can be characterized from the point of view of their suitability as slowing down agents in a chain reaction by a dimensionless constant, N, which we may call the characteristic number. This number, N, is defined by the following formula

$$N = \frac{\sigma_{sc}^*}{\sigma_c} \ln \left( 1 + \frac{2m}{1+m} \right)$$

where m is the mass number of the element,  $\sigma_{sc}^*$  is the scattering cross section of the element for neutrons which are above the thermal region and have energies between a few volts and a few hundred volts, and  $\sigma_c$  is the capture cross section of the element in the thermal region. For pure carbon,  $[\sigma_c(C) \sim 5 \cdot 10^{-27} \text{ cm}^2]$  the characteristic number N(C) is about N(C)  $\sim$  160.

As we have stated before, ~~even~~ impure carbon for which the absorption cross section,  $\sigma_a$  is about twice that of pure carbon is just about capable of sustaining a chain reaction. Such impure carbon would have a characteristic

number of  $N = \frac{1}{2} N(C) = 80$ . Accordingly we may define the class of slowing down agents which are capable of sustaining a chain reaction by the requirement that the characteristic number N should be larger than 80. This class contains heavy hydrogen in its form as deuterium oxide (DO<sub>2</sub>). To compute the approximate value for the characteristic number of deuterium oxide we have to put  $m = 2; \sigma_c = \sigma_c(D) + \frac{1}{2} \sigma_c(O)$ .

The criterion of the characteristic number being larger than about 80 can be applied to all fast neutron scatterers but cannot, without further caution be applied to fast neutron absorbers, i.e., to elements which give with fast fission neutrons an n-p or an n- $\alpha$  reaction, if in the potentially chain reacting system such fast neutrons absorbers are arranged in such a manner as

apart for which the characteristic number is approximately 120 would be sufficient to give this factor. N. We shall call slowing down agents.

This is however no longer possible if we allow the cross section for C to rise appreciably above 10<sup>-26</sup> cm<sup>2</sup>.

preferably be appreciably higher. a value of

to be appreciably exposed to fast fission neutrons. ~~It is for this reason not certain whether beryllium which gives an n- $\alpha$  reaction with fast neutrons can or cannot be used as sole slowing down agent in a potentially chain reacting system, though it appeared to be likely that this is the case.~~

According to the present invention, favorable conditions for a chain reaction may exist in a system which contains the fissionable element within an aggregate of matter which forms the element of a lattice and a lattice of such aggregates is contained in a mass of a suitable, light element which acts as a slowing agent.

The aggregate which forms the lattice element can be a single body containing a <sup>uranium</sup> fissionable element, or can be something like a cluster of a number of separate bodies, each of which contains a fissionable element. This latter case is illustrated by an example shown in Fig. 1. In this figure, we see the lattice element in the form of a spherically shaped aggregate of ~~thin~~ uranium metal rods of different lengths.

The simplest example for a potential chain reacting system is probably one in which the aggregate that forms the lattice element is a single body having the shape of a sphere and being composed of uranium metal. Such uranium metal spheres may then be embedded in graphite in such a manner as to form one of the three existing close-packed lattices. Such a system is potentially chain reacting provided the radius of the uranium metal sphere is between a lower and upper limit which is determined by its density and the ratio of the weight of carbon to uranium within the lattice is kept between certain lower and upper limits which are determined by the radius of the uranium sphere which forms the lattice element.

Lattices

short!  
Fig 1

uranium  
(penetrates)

It is not necessary to use uranium in the form of uranium metal, but  $U_3O_8$  or uranium dioxide or uranium carbide can be used in place of uranium metal. Some other uranium compounds might <sup>perhaps</sup> also be operative.

The aggregate composed of uranium or uranium compounds need not have the shape of a sphere but can have the shape of a short cylinder of about equal diameter and height, ~~(a cube for instance a cube)~~ or can have the shape of a short square rod of about equal sides. <sup>(a body close to a cube)</sup> All these and similar shapes which shall be designated as spheroids are operative, but some other shapes which deviate considerably from the spherical shape are also operative under otherwise favorable conditions.

Lattices in which <sup>is</sup> the aggregate either consists of one uranium-containing body or <sup>is</sup> ~~is~~ composed of several uranium-containing bodies and <sup>in which the aggregate</sup> has the shape of a long cylindrical rod are also operative. Such cylindrical rods can be arranged into a lattice which, for instance, may have trigonal or tetragonal symmetry, but some other lattices which have no such symmetry are also operative.

A recipe will be given further below for an operative close-packed lattice of uranium metal spheres from which recipe an operative combination for the radius of the sphere and the spacing of such spheres in the lattice can be determined. *Can balls?*

Apart from N, the characteristic number of the slowing agent, another property of the slowing agent has a bearing on the question whether the slowing agent is suitable for a system in which a lattice of uranium-containing aggregates is used. For a slowing agent, it is of advantage in this connection that the range, A, of the thermal neutrons

should be large in comparison with range, B, of the low energy resonance neutrons.

A, the range of the thermal neutrons is defined by the value

(3)

$$A = \lambda \sqrt{\frac{\sigma_{sc}}{3\sigma_c}}$$

where  $\lambda$  is the mean free pass of thermal neutrons in the slowing agent, and  $\sigma_x$  and  $\sigma_c$  are the scattering cross-section of the capture cross-section for thermal neutrons of ~~the light element~~ <sup>the molecule</sup> which acts as the slowing agent.

B, the range of the low energy resonance neutrons is defined

by

(4)

$$B = \lambda^* \sqrt{\frac{1}{3}}$$

where  $\lambda^*$  is the mean free pass for scattering of neutrons having the energy between few volts and a few hundred volts in the slowing agent, and  $k$  is defined as follows:

(5)

$$k = \frac{\ln 1/10}{\ln(1 - \frac{2m}{(1+m)^2})}$$

*where m is the mass of the*

Accordingly, the requirements ~~with~~ <sup>that</sup> the range A should be large compared to the range B amounts to

(6)

$$A/B = \frac{\lambda}{\lambda^*} \sqrt{\frac{\sigma_{sc}}{\sigma_c k}} \gg 1$$

or writing  $\frac{\lambda}{\lambda^*} = (\frac{m}{m+1})^2$

(7)

$$A/B = (\frac{m}{m+1})^2 \sqrt{\frac{\sigma_{sc}}{\sigma_c k}} \gg 1$$

This condition is fulfilled for graphite, heavy water, and beryllium, but it is not fulfilled for ordinary water.

*For H2O for instance one has to multiply R by  $\frac{2\sigma_s(D)}{2\sigma_s(D) + \sigma_c(O)}$*

*instead of a pure element (like carbon) and always much heavier element is used as slowing agent. The value given for k has to be multiplied by the fraction of the scattering cross section of the molecule of the compound which is due to the light element.*

If, in stead of an element like carbon, a compound of a light element and a much heavier element is used as slowing agent, the value given for  $k$  has to be multiplied by the fraction of the scattering cross section of the molecule of the compound which is due to the light element.

For D-2, for instance, one has to multiply  $k$  by approximately



For instance, for carbon we have about

$$k \sim 15$$

$$\frac{\sigma_{sc}}{\sigma_c} \sim 1000$$

$$\left(\frac{m}{m+1}\right)^2 = \frac{1}{1.18}$$

*which is satisfactory*  
 This is a ~~very~~ favorable ratio indeed.

Introduction to Graphite Lattice

*before*

For the purposes of this invention, graphite with a low ash content, less than 0.1% ash and preferably less than .06% ash should be used. It is not only important to keep the ash content low, it is also important to keep the boron content of the ash low. If ~~suitable quality~~ *a low boron* petroleum coke is used for making the graphite and if ~~petroleum coke~~ *such* is ~~used for making the graphite and if petroleum coke is also used as~~ *also* ~~the~~ resistor material in the graphitizing process and if the charge in the graphitizing furnace is so located as to reach a very high temperature, one ~~may~~ *can* produce a quality of graphite which is satisfactory.

The uranium compound or uranium metal which is used should also be of high purity. It may be obtained from carnotites or pitchblende by the usual *purification* processes but ~~in addition~~ to these ~~chemical~~ processes, *a* further chemical process of purification should be added, particularly if the uranium is produced from a pitchblende which is comparatively rich in rare earths. This additional process of purification must be so designed as to remove the rare earths from the uranium compound. This may be done by one of several methods,

for instance: by recrystallizations of uranyl nitrate or by extracting uranyl nitrate with ether or by precipitating the rare earths as fluorides, etc. The uranium can be used in the form of  $U_3O_8$ , in the form of the dioxide, in the form of one of its several carbides and in the form of metal. Other uranium compounds might perhaps be also suitable. Uranium metal can be produced from uranium chloride by reduction with calcium and a pure product can be obtained by this method if distilled calcium is used for the process. In a similar manner, pure uranium can be produced by using pure uranium tetrafluoride with chemically pure magnesium. *gm*

~~This latter process is preferable to the former inasmuch as pure magnesium is readily available.~~

*The purest commercial grades of magnesium are substandard.*

## FUNCTIONAL DESCRIPTION

Of the thermal neutrons which are absorbed in the uranium contained in the lattice-element a fraction will cause fission and will give rise to the emission of fast fission neutrons. Some of these fast fission neutrons will cause fission (in the same lattice element from which they originate) before they are slowed down by collisions with uranium or carbon below the fission threshold of the abundant isotope U238. In this manner, for every thermal neutron absorbed a certain number,  $\beta$ , of fast neutrons are generated which are slowed down partly by inelastic collisions in uranium, but mostly by elastic collisions in carbon and a fraction  $(1-\beta)$  of these eventually reaches thermal energies. Another fraction  $\rho$  is absorbed at resonance by uranium before reaching thermal energies.

The neutrons which are absorbed at resonance by uranium are removed from the chain reaction without leading to fission and the generation of neutrons. Some of these neutrons are absorbed at comparatively high energies, between a few hundred volts and some 10,000 volts, while others are absorbed at comparatively low energies, between a few volts and a few hundred volts. The lattice elements are moderately transparent for the high energy resonance neutrons but are practically black for at least part of the low energy resonance neutrons, i.e., a certain fraction of the resonance neutrons is absorbed in a thin surface layer of the lattice element whereas another fraction penetrates. Accordingly, the resonance absorption of the lattice element may be divided into two terms, one of which may be called surface absorption and one of which may be called mass absorption. If the dimensions of the lattice element are small, the fraction of the neutrons which is removed from the chain reaction by mass absorption is essentially determined by the ratio of uranium to carbon and is independent

## Functional Description - 2

of the shape and size of the lattice elements. If it were only for this type of resonance absorption it would be immaterial how small we make the lattice elements and we could make them very small indeed and still have a potentially chain reacting system.

In reality a system in which the dimensions of the lattice element are made very small are not potentially chain reacting since too large a fraction of the neutrons would be removed from the chain reaction by the surface resonance absorption. Clearly if for a given ratio of uranium to carbon we decrease the dimensions of the lattice elements we increase the total surface and thereby the fraction of the neutrons which are removed by absorption at resonance.

In most practical cases a lattice element can be fairly well represented by replacing it with an ellipsoid and we may then express the above-mentioned point of view by saying that the smallest of the three axes of that ellipsoid must not be made too short compared with the range of thermal neutrons in the lattice element. This range "U" is proportionate to the density of the U in the lattice element and increases with the temperature <sup>of the neutrons</sup> somewhat faster than the fourth root of the temperature. For room temperature and uranium metal of density 18 the range is about 142 cm.

INTRODUCTION

According to this invention, a chain reaction can be maintained in a system which contains an element that is capable of undergoing fission under the action of slow neutrons. Such a chain reacting system may be used for the production of radiations of various kinds. Intense neutron radiations are generated and a fraction of the neutrons generated leaves the chain reacting core of the system. Radioactive elements are generated if elements or their compounds are exposed to the neutrons generated in the chain reaction. Of these radioactive elements those produced from uranium or thorium by neutron capture and ~~their daughter~~ <sup>their</sup> ~~products~~ <sup>daughter products</sup> of the substances are of particular interest. Natural uranium containing about one part in 140 of  $U^{235}$  can be used in combination of a ~~slowing down~~ <sup>light</sup> element for building up a chain reacting system. Radioactive elements are then formed by the process of fission both from  $U^{235}$  and also  $U^{238}$ , the latter being induced to fission by means of fast neutrons generated in the fission of  $U^{235}$ . Radioactive <sup>transuranic</sup> elements are also formed by neutron capture from  $U^{238}$  contained in the natural uranium. Energies <sup>is</sup> generated in the form of heat in the fission process and also by the absorption of neutrons and other radiations. <sup>By</sup> means of a cooling agent, the heat produced is led away from the chain reacting system and may be utilized for power production.

~~Further after a period of separation, from separation~~

Of the radioactive elements, which may be generated, of particular interest are the element  $94^{239}$  which is generated from uranium that is exposed

May 14, 1943

to neutrons preferably slow neutrons; and  $U^{233}$  which is generated from thorium. These two elements are of interest because it is believed that they are capable of undergoing fission under the action of slow neutrons and they are, therefore, capable of maintaining a chain reaction if available in sufficient quantities. Of interest also, is polonium which is generated <sup>by neutrons</sup> from bismuth which may be contained in the cooling agent that circulates through the chain reacting system.

In order to be able to separate the radioactive elements from uranium which has been exposed to <sup>slow</sup>neutrons for a certain time in the interior of the chain reacting system, one has to dissolve this uranium. In order to be able to do so, one can proceed in one of two ways:

a. One may remove the uranium from the chain reacting power unit under such precautions as are indicated in view of the strong radioactivity of the uranium, and then dissolve it outside the chain reacting power unit.

b. One may pump a solvent through the power unit and remove the uranium in solution from the chain reaction power unit. If the uranium is present as metal, it may be dissolved either in nitric acid or in a liquid bismuth alloy, and the latter process would have the advantage that no gases are formed within the power unit during the process of solution.

Before removing the uranium from the power unit, it may be advisable to allow the cooling agent <sup>to</sup> circulate through the power unit for a considerable time after the chain reaction has been stopped in order to have the uranium less radioactive at the time of its removal.

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REPLACEMENT FOR SECTION I

According to this invention a nuclear chain reaction can be maintained in a system which contains a lattice of aggregates of a substance that contains an element which is capable of undergoing fission under the action of slow neutrons and which lattice is embedded in a mass <sup>of a suitable</sup> ~~containing at least one~~ *slowing agent.* ~~light element which slows down the fast neutrons emitted during the process~~ of fission of the said heavy element.

Natural uranium is one example of a heavy element that undergoes fission by neutrons which have been slowed down to thermal energies. Fission induced by slow neutrons in natural uranium is attributed to the rare isotope  $U^{235}$  contained in natural uranium. Fast neutrons such as are emitted in the fission of  $U^{235}$  caused, however, fission also in the abundant isotope  $U^{238}$  which is contained in natural uranium. According to this invention a nuclear chain reaction can be maintained in a lattice of aggregates of substance which contains uranium and which is embedded in a mass of a slowing agent which fulfills certain requirements which will be defined further below. Slowing agents which fulfill these requirements will be called, for the purpose of these specifications, efficient slowing agents. Carbon in its pure form of *low ash* graphite belongs to this class of slowing agents and so does deuterium in the form of deuterium oxide, also called heavy water.

Introduction to Graphite Lattice

For the purposes of this invention, graphite with a low ash content, less than 0.1% ash and preferably less than .06% ash should be used. It is not only important to keep the ash content low, it is also important to keep the boron content of the ash low. If a low boron petroleum coke is used for making the graphite and if such petroleum coke is also used as the resistor material in the graphitizing process and if the charge in the graphitizing furnace is

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so located as to reach a very high temperature, one can produce a quality of graphite which is satisfactory.

The uranium compound or uranium metal which is used should also be of high purity. It may be obtained from carnotites or pitchblende by the usual purification processes, but to these processes a further chemical process of purification should be added, particularly if the uranium is produced from a pitchblende which is comparatively rich in rare earths. This additional process of purification must be so designed as to remove the rare earths from the uranium compound. This may be done by one of several methods, for instance: by recrystallizations of uranyl nitrate or by extracting uranyl nitrate with ether or by precipitating the rare earths as fluorides, etc. The uranium can be used in the form of  $U_3O_8$ , in the form of the dioxide, in the form of one of its several carbides and in the form of metal. Other uranium compounds might perhaps be also suitable. Uranium metal can be produced from uranium chloride by reduction with calcium and a pure product can be obtained by this method if distilled calcium is used. In a similar manner, pure uranium can be produced by using pure uranium tetrafluoride with chemically pure magnesium. In this latter process, the purest commercial of magnesium are satisfactory.

Let us now consider an infinitely extended lattice of uranium-containing aggregates embedded in graphite. On the average of the neutrons emitted in a fission process, a fraction  $q$  is absorbed within the system as a slow neutron (thermal or quasi-thermal) and a fraction  $\gamma$  fast neutrons are emitted for every such slow neutron absorbed by uranium. The system is potentially chain reacting if the product  $q\gamma$  is larger than 1.



In order to have an actually chain reacting mass, we must build a sufficiently extended system. If the system is not surrounded with much material which efficiently reflects neutrons, the neutrons can escape from the surface of the chain reacting mass, and in that case, the critical radius  $l$  for which a spherically shaped mass becomes chain reacting, is given by the formula

$$(1) \quad l = \pi \sqrt{\frac{\bar{r}^2}{3(\mu_q - 1)}} \approx \sqrt{\frac{3\bar{r}^2}{\mu_q - 1}}$$

In this formula,  $\bar{r}^2$ , is related to the average square of the distance to which a fast neutron emitted in a fission process diffuses within the system until it gets slowed down and is absorbed and has for graphite of  $1.7 \text{ gm/cm}^3$  of density a value of about  $(50 \text{ cm})^2$  so that we have *in cm for  $l$*

$$l = \sqrt{\frac{7500}{\mu_q - 1}}$$

For a system the composition of which will be described below, and which contains a lattice of aggregates of uranium embedded in graphite of a density of about  $1.6 \text{ gm/cm}^3$ , a sphere containing about 500 tons of pure graphite is sufficiently large to maintain a chain reaction. If in place of a spherical shape, a cylindrical shape is used, and if the diameter of the cylinder is about equal to its height, the critical mass is about 10% higher.

For any given shape the critical mass is defined as the mass at which under the given condition, the intensity of the chain reaction just remains stationary. If the critical mass is exceeded, the intensity of the chain reaction (which manifests itself in the intensity of the neutron and gamma radiations emitted from the chain reacting pile), increases exponentially with time. If the mass is lower than the critical mass, but close to it,

and if we have a source of neutrons (initial neutron radiation) within the mass, the number of neutrons produced in the chain reaction can be made larger than the number of the neutrons of the initial radiation by a factor which increases more and more steeply as the mass approaches to the critical mass. In order to maintain a chain reaction, it is not necessary, however, to make use of an artificial source of neutrons, since initial neutrons are always present; they are for instance emitted in the spontaneous fission of uranium contained in the chain reacting mass and present in the  $\gamma$  rays.

The critical mass nickel or  $\text{Pu}$  can be varied by introducing a slow neutron absorber, such as cadmium or a cobalt iron, or cobalt nickel alloy, into the interior of the mass, or by withdrawing the absorbers therefrom, by which means the critical mass is increased or decreased, respectively. This can be used for controlling the chain reacting, for instance, by moving the slow neutron absorber in and out in such a manner as to have the system part of the time below, and part of the time above. The critical conditions.

In order to determine empirically the critical mass such slow neutron absorbers may be built into the system while the system is being assembled, and after a sufficiently large mass is assembled, these absorbers may be one after the other withdrawn. While these absorbers are being withdrawn, the radiation emanating from the mass may be observed, and the position of the absorbers may be determined at which the neutron intensity begins to rise exponentially with time, which signifies that the critical condition has been reached.

Of the neutrons emitted in fission, a fraction  $\gamma'/\eta$  which is close to 1 is emitted instantaneously and only a small fraction  $1-\gamma'/\eta$  is emitted with a time delay of a few seconds. Though this delayed neutron emission is small, it has an important effect on the rate at which the neutron intensity rises exponentially with time if the mass of the chain reacting unit exceeds the critical

mass, only slightly. In order to discuss this we may introduce the concept of the "critical mass for instantaneous neutron emission" which corresponds to the value of  $M_c$  while the critical mass for the total neutron emission corresponds to  $M_c'$ . The critical mass for instantaneous neutron emission is larger than the critical mass. We can avoid a rapid exponential rise of the chain reaction intensity by keeping conditions so that while we may exceed the critical mass, the system should remain in the interval between the critical mass and the critical mass for instantaneous neutron emission. Preferably in order fully to profit from the effect of the delayed neutron emission, the system should remain within the interval between the two critical masses close to the critical mass for the total neutron emission.

According to the present invention, the chain reaction may be maintained in a system which <sup>contains</sup> is embedded in graphite, a lattice. The lattice element being aggregates of a substance which contains uranium in place of graphite some other efficient slowing agent can be used, for instance, heavy water.

The aggregates which forms the lattice element can be a single body containing uranium or can be something like a cluster of a number of separate bodies, each of which contains uranium. This latter case is illustrated by an example shown in Fig. 1. In this figure we see the lattice element in the form of a spherically shaped aggregate of uranium metal rods (pencils) of different lengths.

The simplest example for a potentially chain reaction system is probably one in which the aggregate that forms the lattice element is a single body having the shape of a sphere and being composed of uranium metal. Such uranium metal spheres may then be embedded in graphite in such a manner as to form one of the three existing close-packed lattices. Such a system is potentially chain reacting provided the radius of the uranium metal sphere

is between a lower and upper limit which is determined by its density and the ratio of the weight of carbon to uranium within the lattice is kept between certain lower and upper limits which are determined by the radius of the uranium sphere which forms the lattice element.

It is not necessary to use uranium in the form of uranium metal, but  $U_3O_8$  or uranium dioxide or uranium carbide can be used in place of uranium metal. Some other uranium compounds might perhaps also be operative.

The aggregate composed of uranium or uranium compounds need not have the shape of a sphere but can have the shape of a short cylinder of about equal diameter and height, or can have the shape of a short ~~cylinder~~ square rod of about equal sides (a body close to a cube). All these and similar shapes which shall be designated as spheroids are operative, but some other shapes which deviate considerably from the spherical shape are operative under otherwise favorable conditions.

Lattices in which is the aggregate either consists of one uranium-containing body or composed of several uranium-containing bodies, and in which the aggregate has the shape of a long cylindrical rod are also operative. Such cylindrical rods can be arranged into a lattice which, for instance, may have trigonal or tetragonal symmetry, but some other lattices which have no such symmetry are also operative.

A recipe will be given further below for an operative close-packed lattice of uranium metal spheres from which recipe an operative combination for the radius of the sphere and the spacing of such spheres in the lattice can be determined.

May 25 43

In the following examples will be given for the following systems:

1. lattices in which the lattice elements are cylindrically shaped uranium rods which may be arranged in trigonal and tetragonal lattice and which are cooled by a cooling agent flowing along the surface of the rod.

Arrangements of this type fall into three classes:

A. a class in which the cooling agent flows inside a uranium tube (insert cooling.)

B. a class in which the cooling agent is flowing along the surface of a uranium rod in a gap between the uranium rod and a thin tube which separates the cooling agent from the slowing agent.

C. arrangements in which the cooling agent flows along the surface of a uranium rod in a gap between the uranium rod and a graphite which acts as a slowing agent.

The class IC is represented by various examples which again fall into two classes, alpha and beta. To the class alpha belongs arrangements in which the cooling agent passes through the whole structure in continuous contact with the lattice element; whereas, to class beta belong arrangements in which various sections of the rod-shaped lattice element are connected in parallel from the point of view of the flow of the cooling agent.

2. lattices of which the elements are spheres of uranium or short cylinders of uranium or uranium compounds of about equal height and diameter. These spheroid-shaped uranium bodies may be arranged in any one of the closed-packed lattices, or may be arranged with a cubic lattice. One or more lattice elements are connected in parallel from the point of view of the flow of the cooling agent.

## INTRODUCTION

According to this invention, a chain reaction can be maintained in a system which contains an element that is capable of undergoing fission under the action of slow neutrons. Such a chain reacting system may be used for the production of radiations of various kinds. Intense neutron radiations are generated and a fraction of the neutrons generated leaves the chain reacting core of the system. Radioactive elements are generated if elements or their compounds are exposed to the neutrons generated in the chain reaction. Of these radioactive elements are produced from uranium or thorium by neutron capture and their daughter products are of particular interest. Natural uranium containing about one part in 140 of  $U^{235}$  can be used in combination of a light element for building up a chain reacting system. Radioactive elements are then formed by the process of fission both from  $U^{235}$  and also  $U^{238}$ , the latter being induced to fission by means of fast neutrons generated in the fission of  $U^{235}$ . Radioactive transuranic elements are also formed by neutron capture from  $U^{238}$  contained in the natural uranium. Energy is generated in the form of heat in the fission process and also by the absorption of neutrons and other radiations. By means of a cooling agent, the heat produced is led away from the chain reacting system and may be utilized for power production.

Of the radioactive elements, which may be generated, of particular interest are the element  $94^{239}$  which is generated from uranium that is exposed to neutrons preferably slow neutrons; and  $U^{233}$  which is generated from thorium. These two elements are of interest because it is believed that they are capable of ~~examined~~ undergoing fission under the action of slow neutrons and they are, therefore, capable of maintaining a chain reaction if available in sufficient quantities. Of interest also, is polonium which is generated by neutrons from bismuth which may be contained in the cooling agent that circulated through the chain reacting system.

In order to be able to separate the radioactive elements from uranium which has been exposed to neutrons for a certain time in the interior of the chain reacting system, one has to dissolve this uranium. In order to be able to do so, one can proceed in one of two ways:

a. One may remove the uranium from the chain reacting power unit under such precautions as are indicated in view of the strong radioactivity of the uranium, and then dissolve it outside the chain reacting power unit.

b. One may pump a solvent through the power unit and remove the uranium in solution from the chain reaction power unit. If the uranium is present as metal, it may be dissolved either in nitric acid or in a liquid bismuth alloy, and the lattice process would have the advantage that no gases are formed within the power unit during the process of solution.

Before removing the uranium from the power unit, it may be advisable to allow the cooling agent to circulate through the power unit for a considerable time after the chain reaction has been stopped in order to have the uranium less radioactive at the time of its removal.

~~III~~ II

May 23, 1943

## REPLACEMENT FOR SECTION I

According to this invention a nuclear chain reaction can be maintained in a system which contains a lattice of aggregates of a substance that contains an element which is capable of undergoing fission under the action of slow neutrons and which lattice is embedded in a mass containing <sup>of a suitable</sup> ~~at least one~~ ~~slowing agent which~~ ~~light element which~~ slows down the fast neutrons emitted during the process of fission of the said heavy element.

Natural uranium is one example of a heavy element that undergoes fission by neutrons which have been slowed down to thermal energies. Fission induced by slow neutrons in natural uranium is attributed to the rare isotope  $U^{235}$  contained in natural uranium. Fast neutrons such as are emitted in the fission of  $U^{235}$  caused, however, fission also in the abundant isotope  $U^{238}$  which is contained in natural uranium. According to this invention a nuclear chain reaction can be maintained in a lattice of aggregates of substance which contains uranium and which is embedded in a mass of a slowing agent which fulfills certain requirements which will be defined further below. Slowing agents which fulfill these requirements will be call<sup>ed</sup>, for the purpose of these specifications, efficient slowing agents. Carbon in its pure form of *low ash* graphite belongs to this class of slowing agents and so does deuterium in the form of deuterium oxide, also called heavy water.

*think*

### Introduction to Graphite Lattice

For the purposes of this invention, graphite with a low ash content, less than 0.1% ash and preferably less than .06% ash should be used. It is not only important to keep the ash content low, it is also important to keep the boron content of the ash low. If a low boron petroleum coke is used for making the graphite and if such petroleum coke is also used as the resistor material in the graphitizing process and if the charge in the graphitizing furnace is



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so located as to reach a very high temperature, one can produce a quality of graphite which is satisfactory.

The uranium compound or uranium metal which is used should also be of high purity. It may be obtained from carnotites or pitchblende by the usual purification processes, but to these processes a further chemical process of purification should be added, particularly if the uranium is produced from a pitchblende which is comparatively <sup>high</sup> ~~rich~~ in rare earths. This additional process of purification must be so designed as to remove the rare earths from the uranium compound. This may be done by one of several methods, for instance: by recrystallizations of uranyl nitrate or by extracting uranyl nitrate with ether or by precipitating the rare earths as fluorides, etc. The uranium can be used in the form of  $U_3O_8$ , in the form of the dioxide, in the form of one of its several carbides and in the form of metal. Other uranium compounds might perhaps be also suitable. Uranium metal can be produced from uranium chloride by reduction with calcium and a pure product can be obtained by this method if distilled calcium is used. In a similar manner, pure uranium can be produced by using pure uranium tetrafluoride with chemically pure magnesium. In this latter process, the purest commercial <sup>brands</sup> of magnesium are satisfactory.

Let us now consider an infinitely extended lattice of uranium-containing aggregates embedded in graphite. On the average of the neutrons emitted in a fission process, a fraction  $q$  is absorbed within the system as a slow neutron (thermal or quasi-thermal) ~~purification of a compound may be solid or liquid,~~ and on the average  $\gamma$  fast neutrons are emitted for every such slow neutron absorbed by uranium. The system is potentially chain reacting if the product  $q\gamma$  is larger than 1.

In order to have an actually chain reacting mass, we must build a sufficiently extended system. If the system is not surrounded with much material which efficiently reflects neutrons, the neutrons can escape from the surface of the chain reacting mass, and in that case, the critical radius  $l$  for which a spherically shaped mass becomes chain reacting, is given by the formula

$$(1) \quad l = \pi \sqrt{\frac{\overline{r^2}}{3(\mu q - 1)}} \approx \sqrt{\frac{3 \overline{r^2}}{\mu q - 1}}$$

In this formula,  $\overline{r^2}$ , is related to the average square of the distance to which a fast neutron emitted in a fission process diffuses within the system until it gets slowed down and is absorbed and has for graphite of 1.7 gm/cm<sup>3</sup> of density a value of about (50 cm)<sup>2</sup> so that we have *in cm for l*

$$l \approx \sqrt{\frac{7500}{\mu q - 1}}$$

For a system the composition of which will be described below, and which contains a lattice of aggregates of uranium embedded in graphite of a density of about 1.6 gm/cm<sup>3</sup>, a sphere containing about 500 tons of pure graphite is sufficiently large to maintain a chain reaction. If in place of a spherical shape, a cylindrical shape is used, and if the diameter of the cylinder is about equal to its height, the critical mass is about 10% higher.

For any given shape the critical mass is defined as the mass at which under the given condition, the intensity of the chain reaction just remains stationary. If the critical mass is exceeded, the intensity of the chain reaction (which manifests itself in the intensity of the neutron and gamma radiations emitted from the chain reacting pile), increases exponentially with time. If the mass is lower than the critical mass, but close to it,

*Wigner*

and if we have a source of neutrons (initial neutron radiation) within the mass, the number of neutrons produced in the chain reaction can be made larger than the number of the neutrons of the initial radiation by a factor which increases more and more steeply as the mass approaches to the critical mass. In order to maintain a chain reaction, it is not necessary, however, to make use of an artificial source of neutrons, since initial neutrons are always present; they are for instance emitted in the spontaneous fission of uranium contained in the chain reacting mass and present in the <sup>atomic</sup> rays.

The critical mass ~~nickel or~~ can be varied by introducing a slow neutron absorber, such as cadmium or <sup>lead or</sup> a cobalt iron, or cobalt nickel alloy, into the interior of the mass, or by withdrawing the absorbers therefrom, by which means the critical mass is increased or decreased, respectively. This can be used for controlling the chain reacting, for instance, by moving the slow neutron absorber in and out in such a manner as to have the system part of the time below, and part of the time above. ~~The~~ critical conditions.

In order to determine empirically the critical mass such slow neutron absorbers may be built into the system while the system is being assembled, and after a sufficiently large mass is assembled, these absorbers may be one after the other withdrawn. While these absorbers are being withdrawn, the radiation emanating from the mass may be observed, and the position of the absorbers may be determined at which the neutron intensity begins to rise exponentially with time, which signifies that the critical condition has been reached.

Of the neutrons emitted in fission, a fraction  $\eta/\eta$  which is close to 1 is emitted instantaneously and only a small fraction  $1-\eta'/\eta$  is emitted with a time delay of a few seconds. Though this delayed neutron emission is small, it has an important effect on the rate at which the neutron intensity rises exponentially with time if the mass of the chain reacting unit exceeds the critical

mass, only slightly. In order to discuss this we may introduce the concept of the "critical mass for instantaneous neutron emission" which corresponds to the value of  $\beta'$  while the critical mass for the total neutron emission corresponds to  $\beta$ . The critical mass for instantaneous neutron emission is larger than the critical mass. We can avoid a rapid exponential rise of the chain reaction intensity by keeping conditions so that while we may exceed the critical mass, the system should remain in the interval between the critical mass and the critical mass for instantaneous neutron emission. Preferably in order fully to profit from the effect of the delayed neutron emission, the system should remain within the interval between the two critical masses close to the critical mass for the total neutron emission.

According to the present invention, <sup>a</sup>the chain reaction may be maintained in a system which <sup>contains</sup> is embedded in graphite, a lattice. The lattice element being aggregates of a substance which contains uranium in place of graphite some other efficient slowing agent can be used, for instance, heavy water.

The aggregates which forms the lattice element can be a single body containing uranium or can be something like a cluster of a number of separate bodies, each of which contains uranium. This latter case is illustrated by an example shown in Fig. 1. This this figure we see the lattice element in the form of a spherically shaped aggregate of uranium metal rods (pencils) of different lengths.

The simplest example for a potentially chain reaction system is probably one in which the aggregate that forms the lattice element is a single body having the shape of a sphere and being composed of uranium metal. Such uranium metal spheres may than be embedded in graphite in such a manner as to form one of the three existing close-packed lattices. Such a system is potentially chain reacting provided the radius of the uranium metal sphere

is between a lower and upper limit which is determined by its density, and the ratio of the weight of carbon to uranium within the lattice is kept between certain lower and upper limits which are determined by the radius of the uranium sphere which forms the lattice element.

It is not necessary to use uranium in the form of uranium metal, but  $U_3O_8$  or uranium dioxide or uranium carbide can be used in place of uranium metal. Some other uranium compounds might perhaps also be operative.

The aggregate composed of uranium or uranium compounds need not have the shape of a sphere but can have the shape of a short cylinder of about equal diameter and height, or can have the shape of a short ~~cylinder~~ square rod of about equal sides (a body close to a cube). All these and similar shapes which shall be designated as spheroids are operative, but some other shapes which deviate considerably from the spherical shape are <sup>also</sup> operative under otherwise favorable conditions.

Lattices in which is the aggregate either consists of one uranium-containing body or composed of several uranium-containing bodies, and in which the aggregate has the shape of a long cylindrical rod are also operative. Such cylindrical rods can be arranged into a lattice which, for instance, may have trigonal or tetragonal symmetry, but some other lattices which have no such symmetry are also operative.

A recipe will be given further below for an operative close-packed lattice of uranium metal spheres from which recipe an operative combination for the radius of the sphere and the spacing of such spheres in the lattice can be determined.

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In the following, examples will be given for the following systems:

1. lattices in which the lattice elements are cylindrically shaped uranium rods which may be arranged in trigonal and tetragonal lattice and which are cooled by a cooling agent flowing along the surface of the rod. Arrangements of this type fall into three classes:

A. a class in which the cooling agent flows inside a uranium tube  
*ide*  
(insert cooling.)

B. a class in which the cooling agent is flowing along the surface of a uranium rod in a gap between the uranium rod and a thin tube which separates the cooling agent from the slowing agent.

C. arrangements in which the cooling agent flows along the surface of a uranium rod in a gap between the uranium rod and a graphite which acts as a slowing agent.

The class iC is represented by various examples which again fall into two classes, alpha and beta. To the class alpha belongs arrangements in which the cooling agent passes through the whole structure in continuous contact with the lattice element; whereas, to class beta belong arrangements in which various sections of the rod-shaped lattice element are connected in parallel from the point of view of the flow of the cooling agent.

2. lattices of which the elements are spheres of uranium or short cylinders of uranium or uranium compounds of about equal height and diameter. These spheroid-shaped uranium bodies may be arranged in any one of the closed-packed lattices, or may be arranged with a cubic lattice. *Groups of* One or more lattice elements are connected in parallel from the point of view of the flow of the cooling agent.

Think

①

According to this invention, a nuclear chain reaction can be maintained in a system which contains a heavy element that undergoes fission under the action of slow neutrons, (such as, for instance, natural uranium) and a light element of a class defined further below, which slows down the fast neutrons emitted during the process of fission by the said heavy element.

Natural uranium is an example for a heavy element that undergoes fission with neutrons that have been slowed down to thermal energies. Fission induced by slow neutrons in natural uranium is attributed to the rare isotope U235, contained in natural uranium. Fast neutrons, such as are emitted in the fission of U235, cause fission also in the abundant isotope, U238, which is contained in natural uranium.

Carbon, particularly in its pure form of low ash graphite, is an example for a light element that can be used for slowing down the neutrons in a potentially chain reacting system. Let us consider an infinitely extended system <sup>of a metal</sup> at a given composition and structure. On the average of the neutrons emitted in a fission process, a fraction  $q$  is absorbed within the system as <sup>slow</sup> neutrons in such a manner as to produce fission, and on the average  $\eta$  fast neutrons are emitted for every fission process induced by slow neutrons. The system is potentially chain reacting if the product  $q\eta$  is larger than 1.

In order to have an actually chain reacting mass, we must build a sufficiently extended system. If the system is not surrounded with much material which efficiently reflects neutrons, the neutrons can escape from the surface of the chain reacting mass, and in that case, the critical radius  $l$  for which a spherically shaped mass becomes chain reacting, is given by the formula

$$l = \pi \sqrt{\frac{F^2}{3(\mu g - 1)}} \sim \sqrt{\frac{3F^2}{\mu g - 1}}$$

In this formula,  $r^2$ , is the average square of the distance to which a fast neutron emitted in a fission process diffuses within the system until it gets slowed down and causes fission.

For a system the composition <sup>of</sup> which will be described below, and which contains a lattice of aggregates of uranium embedded in graphite of a density of about  $1.6 \text{ gm/cm}^3$ , a sphere containing about 500 tons of graphite is sufficiently large to maintain a chain reaction. If in place of a spherical shape, a cylindrical shape is used, and if the diameter of the cylinder is about equal to its height, the critical mass is about 10% higher.

For any given shape the critical mass is defined as the mass at which under the given condition, the intensity of the chain reaction just remains stationery. If the critical mass is exceeded, the intensity of the chain reaction (which manifests itself in the intensity of the neutron for gamma radiation emitted from the chain reacting pile), increases exponentially with time. If the mass is lower than the critical mass, but close to it, and if we have a source of neutrons (initial neutron radiation) within the mass, the number of neutrons produced in the chain reaction can be made larger than the neutrons of the initial radiation as the mass approaches by a factor which increases to infinity to the critical mass. In order to maintain a chain reaction, it is not necessary, however, to make use of an artificial source of neutrons, since initial neutrons are always present; they are for instance emitted in the spontaneous fission of uranium contained in the chain reacting mass.

The critical mass can be varied by introducing a slow neutron absorbers, such as cadmium or a cobalt iron alloy, into the interior of the mass, or by withdrawing the absorbers therefrom, by which means the critical mass is increased or decreased, respectively. This can be used for controlling a chain reaction, for instance, by moving the slow neutron absorber in and out in such a manner as to be part of the time below, and part of the time above, the critical conditions.



In order to determine empirically the critical mass such slow neutron absorbers may be built into the system while the system is being built up, and after a sufficiently large mass is assembled, these absorbers may be one after the other withdrawn. While these absorbers are being withdrawn, the radiation emanating from the mass may be observed, and their position noted at which the neutron intensity begins to rise exponentially with time, which signifies that the critical condition has been reached.

A fraction  $\eta'/\eta$  which is close to 1 is emitted instantaneously and only a small fraction  $1-\eta'/\eta$  is emitted with a time delay of a few seconds. Though this delayed neutron emission is small, it has an important effect on the rate at which the neutron intensity rises exponentially with time if the mass of the chain reacting unit slightly exceeds the critical mass. In order to discuss this we may introduce the concept of the "critical mass for instantaneous neutron emission" which corresponds to the value of  $\eta'$  while the critical mass for the total neutron emission corresponds to  $\eta$ . This critical mass for instantaneous neutron emission is larger than the critical mass. We can avoid a rapid exponential rise of the chain reaction intensity by keeping conditions so that while we may exceed the critical mass, we should remain in the interval between the critical mass and the critical mass for instantaneous fission. Preferably in order fully to profit from the effect of the delayed neutron emission, we should remain within the interval between the two critical masses close to the critical mass for the total neutron emission.

.....

As will be stated further below, a chain reaction can be maintained by means of ordinary uranium in a system in which carbon in the pure form of a low ash graphite is used for slowing down the neutrons. Carbon has an absorption

cross section for thermal neutron of about  $\sigma_c = 5 \times 10^{-27} \text{ cm}^2$ .

If impurities are present, the absorption cross section of the slowing agent per carbon atom is larger and we may allow it to rise perhaps as high as about  $10^{-26} \text{ cm}^2$  before it becomes impossible to maintain a chain reaction in a system of otherwise favorable composition and structure. Carbon and other light elements can be characterized from the point of view of their suitability as slowing down agents in a chain reaction by a dimensionless constant, N, which we may call their characteristic number. This number, N, is defined by the following formula

$$N = \frac{\sigma_{sc}^*}{\sigma_c} \ln \left( 1 + \frac{2m}{1+m^2} \right)$$

where m is the mass number of the element,  $\sigma_{sc}^*$  is the scattering cross section of the element for neutrons which are above the thermal region and have energies between a few volts and a few hundred volts, and  $\sigma_c$  is the capture cross section of the element in the thermal region. For pure carbon,  $(\sigma_c(C) \sim 5 \times 10^{-27} \text{ cm}^2)$  the characteristic number N(C) is about N(C)  $\sim 160$ .

As we have stated before, even impure carbon for which the absorption cross section,  $\sigma_c$  is about twice that of pure carbon is just about capable of sustaining a chain reaction. Such impure carbon would have a characteristic number of  $N = \frac{1}{2} N(C) = 80$ . Accordingly we may define the class of slowing down agents which are capable of sustaining a chain reaction by the requirement that the characteristic number N should be larger than 80. This class contains heavy hydrogen in its form as deuterium oxide ( $\text{D}_2\text{O}$ ). To compute the approximate value for the characteristic number of deuterium oxide we have to put  $m = 2$ ;  $\sigma_c = \sigma_c(D) + \frac{1}{2} \sigma_c(O)$

$\sigma_{sc}^* = \sigma_{sc}^*(D)$  The criterion of the characteristic number being larger than about 80 can be applied to all fast neutron scatterers but cannot, without further caution be applied to fast neutron absorbers, i.e., to elements which gave with fast fission neutrons an n-p or an n- $\alpha$  reaction, if in the potentially chain reacting system such fast neutrons absorbers are arranged in such a manner as

to be appreciably exposed to fast fission neutrons. It is for this reason not certain whether beryllium which gives an  $n-\alpha$  reaction with fast neutrons can or cannot be used as sole slowing down agent in a potentially chain reacting system, though it appeared to be likely that this is the case.

According to the present invention, favorable conditions for a chain reaction may exist in a system which contains the fissionable element within an aggregate of matter which forms the element of a lattice and a lattice of such aggregates is contained in a mass of a suitable, light element which acts as a slowing agent.

The aggregate which forms the lattice element can be a single body containing a fissionable element or can be something like a cluster of a number of separate bodies, each of which contains a fissionable element. This latter case is illustrated by an example shown in Fig. 2. In this figure, we see the lattice element in the form of a spherically shaped aggregate of thin uranium metal rods of different lengths.

The simplest example for a potential chain reacting system is probably one in which the aggregate that forms the lattice element is a single body having the shape of a sphere and being composed of uranium metal. Such uranium metal spheres may then be embedded in graphite in such a manner as to form one of the three existing close-packed lattices. Such a system is potentially chain reacting provided the radius of the uranium metal sphere is between a lower and upper limit which is determined by its density and the ratio of the weight of carbon to uranium within the lattice is kept between certain lower and upper limits which are determined by the radius of the uranium sphere which forms the lattice element.

It is not necessary to use uranium in the form of uranium metal, but  $U_3O_8$  or uranium dioxide or uranium carbide can be used in place of uranium metal. Some other uranium compounds might also be operative.

The aggregate composed of uranium or uranium compounds need not have the shape of a sphere but can have the shape of a short cylinder of about equal diameter and height, or can have the shape of a short square rod of about equal sides. All these and similar shapes which shall be designated as spheroids are operative, but some other shapes which deviate considerably from the spherical shape are also operative under otherwise favorable conditions.

Lattices in which the aggregate either consists of one uranium-containing body or is composed of several uranium-containing bodies and has the shape of a long cylindrical rod are also operative. Such cylindrical rods can be arranged into a lattice which, for instance, may have trigonal or tetragonal symmetry, but some other lattices which have no such symmetry are also operative.

A recipe will be given further below for an operative close-packed lattice of uranium metal spheres from which recipe an operative combination for the radius of the sphere and the spacing of such spheres in the lattice can be determined.

Apart from  $N$ , the characteristic number of the slowing agent, another property of the slowing agent has a bearing on the question whether the slowing agent is suitable for a system in which a lattice of uranium-containing aggregates is used. For a slowing agent, it is of advantage in this connection that the range,  $A$ , of the thermal neutrons

should be large in comparison with range, B, of the low energy resonance neutrons.

A, the range of the thermal neutrons is defined by the value

$$A = \lambda \sqrt{\frac{\sigma_{sc}}{3 \sigma_c}}$$

where  $\lambda$  is the mean free pass of thermal neutrons in the slowing agent, and  $\sigma_{sc}$  and  $\sigma_c$  are the scattering cross-section of the capture cross-section for thermal neutrons of the light element which acts as the slowing agent.

B, the range of the low energy resonance neutrons is defined by

$$B = \lambda^* \sqrt{\frac{k}{3}}$$

where  $\lambda^*$  is the mean free pass for scattering of neutrons having the energy between few volts and a few hundred volts in the slowing agent, and k is defined as follows:

$$k = \frac{\ln 1/10}{\ln(1 - \frac{2m}{(1+m)^2})}$$

Accordingly, the requirements with the range A should be large compared to the range B amounts to

$$A/B = \frac{\lambda}{\lambda^*} \sqrt{\frac{\sigma_{sc}}{\sigma_c k}} \gg$$

or writing  $\frac{\lambda}{\lambda^*} = \left(\frac{m}{m+1}\right)^2$

$$A/B = \left(\frac{m}{m+1}\right)^2 \sqrt{\frac{\sigma_{sc}}{\sigma_c k}}$$

This condition is fulfilled for graphite, heavy water, and beryllium, but it is not fulfilled for ordinary water.

For instance, for carbon we have about

$$k \sim 15$$

$$\frac{\sigma_{sc}}{\sigma_c} \sim 1000$$

$$\left(\frac{m}{m+1}\right)^2 = \frac{1}{1.18}$$

$$A/B \sim 7$$

This is a very favorable ratio indeed.

#### Introduction to Graphite Lattice

For the purposes of this invention, graphite with a low ash content, less than 0.1% ash and preferably less than .06% ash should be used. It is not only important to keep the ash content low, it is also important to keep the boron content of the ash low. If suitable quality of petroleum coke is used for making the graphite and if petroleum coke is used for making the graphite and if petroleum coke is also used as a resistor material in the graphitizing process and if the charge in the graphitizing furnace is so located as to reach a very high temperature, one may produce a quality of graphite which is satisfactory.

The uranium compound or uranium metal which is used should also be of high purity. It may be obtained from carnotites or pitchblende by the usual processes but in addition to these chemical processes, further chemical process of purification should be added, particularly if the uranium is produced from a pitchblende which is comparatively rich in rare earths. This additional process of purification must be so designed as to remove the rare earths from the uranium compound. This may be done by one of several methods,

for instance: by recrystallizations of uranyl nitrate or by extracting uranyl nitrate with ether or by precipitating the rare earths as fluorides, etc. The uranium can be used in the form of  $U_3O_8$ , in the form of the dioxide, in the form of one of its several carbides and in the form of metal. Other uranium compounds might perhaps be also suitable. Uranium metal can be produced from uranium chloride by reduction with calcium and a pure product can be obtained by this method if distilled calcium is used for the process. In a similar manner, pure uranium can be produced by using pure uranium tetrafluoride with chemically pure magnesium. This latter process is preferable to the former inasmuch as pure magnesium is readily available.

## FUNCTIONAL DESCRIPTION

Of the thermal neutrons which are absorbed in the uranium contained in the lattice-element a fraction will cause fission and will give rise to the emission of fast fission neutrons. Some of these fast fission neutrons will cause fission (in the same lattice element from which they originate) before they are slowed down by collisions with uranium or carbon below the fission threshold of the abundant isotope U238. In this manner, for every thermal neutron absorbed a certain number of fast neutrons are generated which are slowed down partly by inelastic collisions in uranium, but mostly by elastic collisions in carbon and a fraction ( ) of these eventually reaches thermal energies. Another fraction is absorbed at resonance by uranium before reaching thermal energies.

The neutrons which are absorbed at resonance by uranium are removed from the chain reaction without leading to fission and the generation of neutrons. Some of these neutrons are absorbed at comparatively high energies, between a few hundred volts and some 10,000 volts, while others are absorbed at comparatively low energies, between a few volts and a few hundred volts. The lattice elements are moderately transparent for the high energy resonance neutrons but are practically black for at least part of the low energy resonance neutrons, i.e., a certain fraction of the resonance neutrons is absorbed in a thin surface layer of the lattice element whereas another fraction penetrates. Accordingly, the resonance absorption of the lattice element may be divided into two terms, one of which may be called surface absorption and one of which may be called mass absorption. If the dimensions of the lattice element are small, the fraction of the neutrons which is removed from the chain reaction by mass absorption is essentially determined by the ratio of uranium to carbon and is independent



Functional Description - 2

of the shape and size of the lattice elements. If it were only for this type of resonance absorption it would be immaterial how small we make the lattice elements and we could make them very small indeed and still have a potentially chain reacting system.

In reality a system in which the dimensions of the lattice element are made very small are not potentially chain reacting since too large a fraction of the neutrons would be removed from the chain reaction by the surface resonance absorption. Clearly if for a given ratio of uranium to carbon we decrease the dimensions of the lattice elements we increase the total surface and thereby the fraction of the neutrons which are removed by absorption at resonance.

In most practical cases a lattice element can be fairly well represented by replacing it with an ellipsoid and we may then express the above-mentioned point of view by saying that the smallest of the three axes of that ellipsoid must not be made too short compared with the range of thermal neutrons in the lattice element. This range "U" is proportionate to the density of the U in the lattice element and increases with the temperature somewhat faster than the fourth root of the temperature. For room temperature and uranium metal of density 18 the range is about 142 cm.

## I

## INTRODUCTION

According to this invention, a chain reaction can be maintained in a system which contains an element that is capable of undergoing fission under the action of slow neutrons. Such a chain reacting system may be used for the production of radiations of various kinds. Intense neutron radiations are generated and a fraction of the neutrons generated leaves the chain reacting core of the system. Radioactive elements are generated if elements or their compounds are exposed to the neutrons generated in the chain reaction. Of these, radioactive elements produced from uranium or thorium by neutron capture and their daughter products are of particular interest.

Natural uranium containing about one part in 140 of  $U^{235}$  can be used in combination of a light element for building up a chain reacting system. Radioactive elements are then formed by the process of fission both from  $U^{235}$  and also  $U^{238}$ , the latter being induced to fission by means of fast neutrons generated in the fission of  $U^{235}$ . Radioactive transuranic elements are also formed by neutron capture from  $U^{238}$  contained in the natural uranium. Energy is generated in the form of heat in the fission process and also by the absorption of neutrons and other radiations. By means of a cooling agent, the heat produced is led away from the chain reacting system and may be utilized for power production.

Of the radioactive elements, which may be generated, of particular interest are the element  $94^{239}$  which is generated from uranium that is exposed to neutrons preferably slow neutrons; and  $U^{233}$  which is generated from thorium. These two elements are of interest because it is believed that they are capable of undergoing fission under the action of slow

neutrons and they are, therefore, capable of maintaining a chain reaction if available in sufficient quantities. Of interest also, is polonium which is generated by neutrons from bismuth which may be contained in the cooling agent that circulates through the chain reacting system.

In order to be able to separate the radioactive elements from uranium which has been exposed to neutrons for a certain time in the interior of the chain reacting system, one has to dissolve this uranium. In order to be able to do so, one can proceed in one of two ways:

a. One may remove the uranium from the chain reacting power unit under such precautions as are indicated in view of the strong radioactivity of the uranium, and then dissolve it outside the chain reacting power unit.

b. One may pump a solvent through the power unit and remove the uranium in solution from the chain reaction power unit. If the uranium is present as metal, it may be dissolved either in nitric acid or in a liquid bismuth alloy, and the latter process would have the advantage that no gases are formed within the power unit during the process of solution.

Before removing the uranium from the power unit, it may be advisable to allow the cooling agent to circulate through the power unit for a considerable time after the chain reaction has been stopped in order to have the uranium less radioactive at the time of its removal.

May 23, 1943

## II

According to this invention a nuclear chain reaction can be maintained in a system which contains a lattice of aggregates of a substance that contains an element which is capable of undergoing fission under the action of slow neutrons and which lattice is embedded in a mass containing a suitable slowing agent which slows down the fast neutrons emitted during the process of fission of the said heavy element.

Natural uranium is one example of a heavy element that undergoes fission by neutrons which have been slowed down to thermal energies. Fission induced by slow neutrons in natural uranium is attributed to the rare isotope  $U^{235}$  contained in natural uranium. Fast neutrons such as are emitted in the fission of  $U^{235}$  cause, however, fission also in the abundant isotope  $U^{238}$  which is contained in natural uranium. According to this invention a nuclear chain reaction can be maintained in a lattice of aggregates of substance which contains uranium and which is embedded in a mass of a slowing agent which fulfills certain requirements which will be defined further below. Slowing agents which fulfill these requirements will be called, for the purpose of these specifications, "efficient" slowing agents. Carbon in its pure form of low ash graphite belongs to this class of slowing agents and so does deuterium in the form of deuterium oxide, also called heavy water.

Introduction to Graphite Lattice.

For the purposes of this invention, graphite with a low ash content, less than 0.1% ash and preferably less

than 06% ash should be used. It is not only important to keep the ash content low, it is also important to keep the boron content of the ash low. If a low boron petroleum coke is used for making the graphite and if such petroleum coke is also used as the resistor material in the graphitizing process and if the charge in the graphitizing furnace is so located as to reach a very high temperature, one can produce a quality of graphite which is satisfactory.

The uranium compound or uranium metal which is used should also be of high purity. It may be obtained from carnotites or pitchblende which is comparatively high in rare earths. This additional process of purification must be so designed as to remove the rare earths from the uranium compound. This may be done by one of several methods, for instance; by recrystallization of uranyl nitrate or by extracting uranyl nitrate with ether or by precipitating the rare earths as fluorides, etc..

The uranium can be used in the form of  $U_3O_8$ , in the form of the dioxide, in the form of one of its several carbides and in the form of metal. Other uranium compounds might perhaps be also suitable. Uranium metal can be produced from uranium chloride by reduction with calcium and a pure product can be obtained by this method if distilled calcium is used. In a similar manner, pure uranium can be produced by using pure uranium tetrafluoride with chemically pure magnesium. In this latter process, the purest commercial brands of magnesium are satisfactory.

Let us now consider an infinitely extended lattice of uranium-containing aggregates embedded in graphite. On

the average of the neutrons emitted in a fission process, a fraction  $q$  is absorbed within the system as a slow neutron (thermal or quasi-thermal) by uranium, and on the average  $\eta$  fast neutrons are emitted for every such slow neutron absorbed by uranium. The system is potentially chain reacting if the product  $q\eta$  is larger than 1.

In order to have an actually chain reacting mass, we must build a sufficiently extended system. If the system is not surrounded with much material which efficiently reflects neutrons, the neutrons can escape from the surface of the chain reacting mass, and in that case, the critical radius  $l$  for which a spherically shaped mass becomes chain reacting, is given by the formula

$$(1) \quad l = \pi \sqrt{\frac{r^2}{3(kg-1)}} \approx \sqrt{\frac{3kr^2}{kg-1}}$$

In this formula,  $r^2$ , is related to the average square of the distance to which a fast neutron emitted in a fission process diffuses within the system until it gets slowed down and is absorbed and has for graphite of 1.7 gm/cm<sup>3</sup> of density a value of about (50 cm)<sup>2</sup> so that we have

$$l \approx \sqrt{\frac{7500}{kg-1}}$$

For a system the composition of which will be described below, and which contains a lattice of aggregates of uranium embedded in graphite of a density of about 1.6 gm/cm<sup>3</sup>, a sphere containing about 500 tons of pure graphite is sufficiently large to maintain a chain reaction. If in place of a spherical shape, a cylindrical shape is used, and if the diameter of the cylinder is about equal to its height, the critical mass is about

10% higher.

For any given shape the critical mass is defined as the mass at which under the given condition, the intensity of the chain reaction just remains stationary. If the critical mass is exceeded, the intensity of the chain reaction (which manifests itself in the intensity of the neutron and gamma radiations emitted from the chain reacting pile), increases exponentially with time. If the mass is lower than the critical mass, but close to it, and if we have a source of neutrons (initial neutron radiation) within the mass, the number of neutrons produced in the chain reaction can be made larger than the number of the neutrons of the initial radiation by a factor which increases more and more steeply as the mass approaches to the critical mass. In order to maintain a chain reaction, it is not necessary, however, to make use of an artificial source of neutrons, since initial neutrons are always present; they are for instance emitted in the spontaneous fission of uranium contained in the chain reacting mass and present in the cosmic rays.

The critical mass can be varied by introducing a slow neutron absorber, such as cadmium, steel, a cobalt iron, or cobalt nickel alloy, into the interior of the mass, or by withdrawing the absorbers therefrom, by which means the critical mass is increased or decreased, respectively. This can be used for controlling the chain reacting, for instance, by moving the slow neutron absorber in and out in such a manner as to have the system part of the time below, and part of the time above, the critical condition.

In order to determine empirically the critical mass such slow neutron absorbers may be built into the system

while the system is being assembled, and after a sufficiently large mass is assembled, these absorbers may be one after the other withdrawn. While these absorbers are being withdrawn, the radiation emanating from the mass may be observed, and the position of the absorbers may be determined at which the neutron intensity begins to rise exponentially with time, which signifies that the critical condition has been reached.

Of the neutrons emitted in fission, a fraction  $\eta/\eta$  which is close to 1 is emitted instantaneously and only a small fraction  $1 - \eta/\eta$  is emitted with a time delay of a few seconds. Though this delayed neutron emission is small, it has an important effect on the rate at which the neutron intensity rises exponentially with time if the mass of the chain reacting unit exceeds the critical mass, only slightly. In order to discuss this we may introduce the concept of the "critical mass for instantaneous neutron emission" which corresponds to the value of  $\beta'$  while the critical mass for the total neutron emission corresponds to  $\beta$ . The critical mass for instantaneous neutron emission is larger than the critical mass. We can avoid a rapid exponential rise of the chain reaction intensity by keeping conditions so that while we may exceed the critical mass, the system should remain in the interval between the critical mass and the critical mass for instantaneous neutron emission. Preferably in order fully to profit from the effect of the delayed neutron emission, the system should remain within the interval between the two critical masses close to the critical mass for the total neutron emission.

According to the present invention, a chain reaction may be maintained in a system which contains embedded in graphite, a lattice, the lattice element being aggregates



of a substance which contains uranium, in place of graphite some other efficient slowing agent can be used, for instance, heavy water.

The aggregate which forms the lattice element can be a single body containing uranium or can be something like a cluster of a number of separate bodies, each of which contains uranium. This latter case is illustrated by an example shown in Fig. 1. This figure we see the lattice element in the form of a spherically shaped aggregate of uranium metal rods (pencils) of different lengths.

The simplest example for a potentially chain reaction system is probably one in which the aggregate that forms the lattice element is a single body having the shape of a sphere and being composed of uranium metal. Such uranium metal spheres may then be embedded in graphite in such a manner as to form one of the three existing close-packed lattices. Such a system is potentially chain reacting provided the radius of the uranium metal sphere is between a lower and upper limit which is determined by its density, and the ratio of the weight of carbon to uranium within the lattice is kept between certain lower and upper limits which are determined by the radius of the uranium sphere which forms the lattice element.

It is not necessary to use uranium in the form of uranium metal, but  $U_3O_8$  or uranium dioxide or uranium carbide can be used in place of uranium metal. Some other uranium compounds might perhaps also be operative.

The aggregate composed of uranium or uranium compounds need not have the shape of a sphere but can have the shape of a short cylinder of about equal diameter and height, or can have the shape of a short square rod of about equal sides (a body close to a cube). All these and similar

shapes which shall be designated as spheroids are operative, but some other shapes which deviate considerably from the spherical shape are also operative under otherwise favorable conditions.

Lattices in which the aggregate either consists of one uranium-containing body or composed of several uranium-containing bodies, and in which the aggregate has the shape of a long cylindrical rod are also operative. Such cylindrical rods can be arranged into a lattice which, for instance, may have trigonal or tetragonal symmetry, but some other lattices which have no such symmetry are also operative.

A recipe will be given further below for an operative close-packed lattice of uranium metal spheres from which recipe an operative combination for the radius of the sphere and the spacing of such spheres in the lattice can be determined.

In the following, examples will be given for the following systems:

1. Lattices in which the lattice elements are cylindrically shaped uranium rods which may be arranged in trigonal and tetragonal lattice and which are cooled by a cooling agent flowing along the surface of the rod. Arrangements of this type fall into three classes:

A. A class in which the cooling agent flows inside a uranium tube (inside cooling).

B. A class in which the cooling agent is flowing along the surface of a uranium rod in a gap between the uranium rod and a thin tube which separates the cooling agent from the slowing agent.

C. Arrangements in which the cooling agent flows along the surface of a uranium rod in a gap between the uranium rod and graphite which acts as a slowing agent.

The class 1C is represented by various examples which again fall into two classes, alpha and beta. To the class alpha belongs arrangements in which the cooling agent passes through the whole structure in continuous contact with the lattice element; whereas, to class beta belong arrangements in which various sections of the rod-shaped lattice element are connected in parallel from the point of view of the flow of the cooling agent.

2. Lattices of which the elements are spheres of uranium or short cylinders of uranium or uranium compounds of about equal height and diameter. These spheroid-shaped uranium bodies may be arranged in any one of the closed-packed lattices, or may be arranged in a cubic lattice. Groups of one or more lattice elements are connected in parallel from the point of view of the flow of the cooling agent.

## INTRODUCTION

According to this invention, a chain reaction can be maintained in a system which contains an element that is capable of undergoing fission under the action of slow neutrons. Such a chain reacting system may be used for the production of radiations of various kinds. Intense neutron radiations are generated and a fraction of the neutrons generated leaves the chain reacting core of the system. Radioactive elements are generated if elements or their compounds are exposed to the neutrons generated in the chain reaction. Of these, radioactive elements produced from uranium or thorium by neutron capture and their daughter products are of particular interest. Natural uranium containing about one part in 140 of  $U^{235}$  can be used in combination of a light element for building up a chain reacting system. Radioactive elements are then formed by the process of fission both from  $U^{235}$  and also  $U^{238}$ , the latter being induced to fission by means of fast neutrons generated in the fission of  $U^{235}$ . Radioactive transuranic elements are also formed by neutron capture from  $U^{238}$  contained in the natural uranium. Energy is generated in the form of heat in the fission process and also by the absorption of neutrons and other radiations. By means of a cooling agent, the heat produced is led away from the chain reacting system and may be utilized for power production.

Of the radioactive elements, which may be generated, of particular interest are the element  $94^{239}$  which is generated from uranium that is exposed to neutrons, preferably slow neutrons; and  $U^{233}$  which is generated from thorium. These two elements are of interest because it is believed that they are capable of undergoing fission under the action of slow

neutrons and they are, therefore, capable of maintaining a chain reaction if available in sufficient quantities. Of interest also, is polonium which is generated by neutrons from bismuth which may be contained in the cooling agent that circulates through the chain reacting system.

In order to be able to separate the radioactive elements from uranium which has been exposed to neutrons for a certain time in the interior of the chain reacting system, one has to dissolve this uranium. In order to be able to do so, one can proceed in one of two ways:

a. One may remove the uranium from the chain reacting power unit under such precautions as are indicated in view of the strong radioactivity of the uranium, and then dissolve it outside the chain reacting power unit.

b. One may pump a solvent through the power unit and remove the uranium in solution from the chain reaction power unit. If the uranium is present as metal, it may be dissolved either in nitric acid or in a liquid bismuth alloy, and the latter process would have the advantage that no gases are formed within the power unit during the process of solution.

Before removing the uranium from the power unit, it may be advisable to allow the cooling agent to circulate through the power unit for a considerable time after the chain reaction has been stopped in order to have the uranium less radioactive at the time of its removal.

I  
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Before removing the uranium from the power unit, it may be advisable to allow the cooling agent to circulate through the power unit for a considerable time after the chain reaction has been stopped in order to have the uranium less radioactive at the time of its removal.