DIVERGENT CHAIN REACTION IN SYSTEMS COMPOSED

OF URANIUM AND CARBON

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In July 1939, it became first apparent to the author that there is a serious possibility of maintaining a chain reaction in a system composed of uranium and graphite. An experiment completed in collaboration with H. L. Anderson and E. Fermi, earlier in June 1939, had reliably indicated that a water-uranium system can indeed come reasonably close to be chain reacting. At that time there was only very imperfect knowledge of the values of the nuclear constants involved but this did not stand in the way of making a comparison between a heterogeneous uranium-carbon system and a homogeneous uranium-water system. The comparison showed that if a homogeneous water-uranium system can be constructed which comes very close to be chain reacting, then it should be possible to make a heterogeneous carbon-uranium system chain reacting, provided that the absorption of carbon is lower than $.01 \times 10^{-24}$, which happened to be the experimental upper limit for carbon absorption at that time. The Government was advised of this situation in October 1939.

No further experiments on such systems were undertaken at Columbia University, or for that matter anywhere in the United States, between June 1939 and March 1940. But in January 1940, an experiment performed by Halban, Joliot, Kowarski, and Perrin, on uranium-water systems became known in this country. One of the uranium-water systems investigated by them came close to be capable of maintaining a chain reaction and one could also see that such systems could get exceedingly close to be chain reacting for the optimum concentration. In the opinion of the author, this meant that we can expect a chain reaction in a uranium-carbon system under practically attainable conditions, if the capture cross-section of carbon had a value of, say, about one half of the experimental upper limit quoted above, i.e., .005.

Thus it appeared important to try to obtain a rough idea as to the optimal composition, geometrical dimensions, and other characteristics of such a chain reacting system and this was attempted in the present paper. Over six years have now elapsed since it was written and naturally it is outdated. A few notes were added to draw attention to some of the shortcomings which have become evident in the meantime. Some parts of the paper have been condensed or left out entirely in an attempt to shorten it without adding anything to its original contents.

INTRODUCTION

As early as 1913, H. G. Wells forecast the discovery of induced radioactivity for the year 1933 and described the subsequent advent of nuclear transmutations on an industrial scale. It was not possible for physicists to realize the potentialities of nuclear physics in this direction until after the discovery of the neutron in 1932. Owing to the discovery of artificial radio-activity in 1933 by Joliot and Irene Curie, and Fermi's pioneer work on neutron reactions in 1934, progress in the

field of nuclear physics was greatly accelerated. By the middle of 1934 it could be clearly seen that within the framework of modern nuclear physics transmutation of elements on an industrial scale might be achieved by means of a chain reaction in which neutrons form the links of the chain. If there is an element or a mixture of elements which interacts with neutrons and from which a neutron liberates on the average more than one neutron for one neutron which is absorbed within the mixture we have a chain reaction and may bring about nuclear transmutations on a large scale. If a neutron source is placed in the center of a large sphere which is composed of such an element or mixture the number of neutrons emerging out of the sphere will be larger than the number of neutrons emitted by the source in the center of the sphere. If the radius of the sphere approached a certain critical radius the number of neutrons generated in the chain would tend to become infinite. If the radius of the sphere is larger than the critical radius no stationary state is possible and the number of neutrons would increase exponentially with time.

By simplifying the problem so as to be able to apply the theory of diffusion to the motion of the neutrons which are liberated in the chain reaction it is easy to get an approximate picture of the general type of behaviour of a chain reaction within a finite sphere of matter. Such a simplified treatment has been applied to the problem in 1934, particularly with regard to the potential possibility of setting up a chain reaction [*20] in beryllium. Later this idea had to be abandoned, as far as beryllium was concerned, when Aston's value for the mass of helium proved to be in error.

In 1934 the transmutation of uranium by neutrons was discovered along that of other elements by Amaldi, D' Agostino, Fermi, Rasetti, and Segre who found that a number of radioactive elements are generated from uranium by neutrons. An important advance was made by Irene Curie and P. Savitch, who found that an element which behaved chemically apparently like radium was among those produced and later Irene Curie and P. Savitch discovered radioactive rare earths among the disintegration products of uranium. Finally Hahn and Strassman announced in December 1938, that in reality uranium splits into elements of medium atomic weight if irradiated by neutrons.

The possibility of a chain reaction was indicated by the discovery of a strong instantaneous emission of neutrons by uranium under the action of thermal neutrons. This was found independently and almost practically simultaneously, by Halban, Joliot, and

(3)   (4)   (5)
Kowarski ; Anderson and Fermi; and Szilard and Zinn who reported their observations

in letters to <u>Nature</u> and to <u>Physical Review</u> dated March 8, March 16, and March 16 (1939)

respectively. An observation pointing in the same direction was also reported by
      (6)
v. Droste   in a letter to <u>Die Naturwissenschaften</u> dated March 17, (1939).

The balance of neutron absorption and emission by uranium was subsequently
                          (11)   (12)
studied by Halban, Joliot and Kowarski; Anderson, Fermi and Szilard; and Halban,
                  (13)
Joliot, Kowarski, and Perrin.   These experiments showed on the average, more than

one fast neutron is emitted for one thermal neutron absorbed by uranium. They show,

therefore, that in certain circumstances a chain reaction might be maintained in a

mixture of uranium and an element which slows down to thermal energies the fast neut-

rons emitted by the splitting uranium. If it is possible to avoid that too large a

fraction of the fast neutrons emitted be captured without causing fission by uranium

at resonance and if it is possible at the same time to avoid that too large a fraction

of the thermal neutrons be captured by the element which is used for the purpose of

slowing down, we would have a chain reaction which may diverge for a sufficiently

large mass of uranium.

This does not necessarily mean that a chain reaction can be maintained

in a mixture of water and uranium oxide. Fermi has investigated (June 1939) the

question whether more favorable conditions can be obtained in mixtures of uranium

oxide and water by keeping the uranium oxide and water in separate layers and found

that a slight improvement can thus be obtained. But this improvement was not sufficient

to determine whether a chain reaction can be obtained in a system composed of water

and uranium oxide.

A recent experiment published by Halban, Joliot, Kowarski, and Perrin,
                          (13)
on homogeneous uranium-water systems will be discussed below and it will be seen

that such systems, while they may or may not be actually capable of sustaining a

chain reaction, can in any case come very close to it.

The Graphite-Uranium System

The purpose of the present paper is to show that a chain reaction can be achieved by using an element like carbon for slowing down the neutrons in certain particular systems composed of uranium and carbon. The theory which is given in the present paper can be applied to elements other than carbon but it does not give any useful information for systems composed of uranium and hydrogen.

Though one might think that carbon should be much less efficient for slowing down neutrons than hydrogen, from several points of view it would be preferable to use carbon in the form of graphite rather than hydrogen in the form of water. The capture cross section of carbon for thermal neutrons $\sigma_c(C)$ is small. An upper limit of $\sigma_c(C) < 0.01$ has been reported by Frisch, Halban and Koch, but this upper limit is not sufficiently low to allow us at present to conclude that a chain reaction could be maintained in homogeneous mixtures of uranium and carbon. For neutrons it takes about 6.5 collisions with carbon atoms to reduce their energy by a factor of e. [16] Thus a neutron which is being slowed down by carbon stays for a long time within the resonance absorption region of uranium. Consequently, very low uranium concentrations would have to be used in order to avoid that a large fraction of the fast neutrons emitted by uranium is absorbed at resonance by uranium. At such very low uranium concentrations, on the other hand, the fraction of the thermal neutrons which is absorbed by carbon might perhaps be too large to permit a chain reaction.

It will be shown, however, in the present paper that if instead of using a homogeneous mixture of uranium and carbon, a large number of spheres of uranium which may form, for instance, a close-packed hexagonal or cubic lattice are embedded in carbon, the ratio of the number of thermal neutrons and the number of resonance neutrons absorbed by the uranium can be so much increased that a chain reaction will become possible. [*21] It will be seen that this ratio strongly depends on the radius of the uranium spheres and that a rather small radius must be chosen in order to obtain most favorable conditions.

SINGLE SPHERE OF URANIUM

We wish to first calculate $\mathcal{E}$, the ratio of the number of thermal neutrons and the number of resonance neutrons which are absorbed in a single uranium sphere under the following conditions: The sphere is embedded in an infinite space filled with carbon in which there is a uniform production of resonance neutrons throughout the whole mass and a uniform production of thermal neutrons throughout the whole mass. For the number of resonance neutrons and thermal neutrons produced per cubic centimeter and second is taken to be equal. In order to obtain an estimate for the value of $\mathcal{E}$ we shall assume that all neutrons which have an energy between some lower energy E_1, and another higher energy $E_2 = 10\ E_1$ are absorbed by uranium at resonance if they reach the surface of the uranium sphere by diffusion.[*]

A neutron which is slowed down by elastic collisions with carbon atoms and which enters the resonance at $E = E_2$ will survive on the average k^{res} collisions within the resonance region between E_2 and E_1 and we have

$$k^{res} = 6.5\ \ln \frac{E_2}{E_1}$$

Under the assumption which we have made above, i.e., $E_2 = 10\ E_1$

$$k^{res} = 6.5\ \ln 10 = 15$$

This may be compared with the average number of elastic collisions k^{th} which a thermal neutron will survive in carbon before being captured by a carbon atom. Since the capture cross-section of carbon is small compared with σ_{sc} the scattering cross-section of carbon for thermal neutrons the probability $g_1(k)$ that a thermal neutron will survive collisions in carbon will be given by

$$g_1(k) = e^{k\ \frac{\sigma_c}{\sigma_{sc}}}$$

Accordingly, k^{th} the average number of collisions which a thermal neutron will make with carbon atoms before being captured is given by

$$k^{th} = \int_0^{\infty} k\ g_1(k)\ dk$$

so that

$$k^{th} = \frac{\sigma_{sc}}{\sigma_c}$$

Let us now first determine the number of thermal neutrons which are absorbed by a single uranium sphere of radius R embedded in an infinite space filled with carbon if Q thermal neutrons are produced per c.c. and second in the carbon. If R is large compared to λ the mean free path for elastic scattering of thermal neutrons in carbon, the density ρ of the thermal neutrons in the carbon can be calculated by treating the problem as a diffusion phenomenon. We thus find for ρ as a function of the distance r from the center of the sphere

$$(3) \qquad D \frac{d(r\rho)}{dr^2} - S\,r\rho + Q(r)\,r = 0$$
$$D = \frac{v\lambda}{3} \; ; \; S = \frac{v}{\lambda}\frac{\sigma_c}{\sigma_{sc}}$$

If the same number of thermal neutrons are produced everywhere in the carbon per c.c. and second we have

$$(4) \qquad \frac{dQ}{dr} = 0$$

For a sphere which absorbs each thermal neutron which reaches its surface i.e. for a "black" sphere we have $\rho(R) = 0$ and find from (3) and (4) for $r > R$

$$(5) \qquad \rho(r) = \frac{Q}{S}\left(1 - \frac{R}{r}\,e^{-\frac{r-R}{A}}\right)$$

$$(6) \qquad A = \sqrt{\frac{D}{S}} = \frac{\lambda}{\sqrt{3}}\sqrt{\frac{\sigma_{sc}}{\sigma_c}} = \frac{\lambda}{\sqrt{3}}\sqrt{k^{u}}$$

the number of thermal neutrons which is absorbed by a single uranium sphere per second is given by

$$(7) \qquad J^{th} = D\cdot 4\pi R^2 \rho'(R)$$

and for a black uranium sphere we find from No. 5 $\qquad J^{th} = J_o^{u}$

$$(8) \qquad J_o^{u} = 4\pi Q\,R A^2\left(1 + \frac{R}{A}\right) \; ; \; A = \frac{\lambda}{\sqrt{3}}\sqrt{k^{u}}$$

where A has the dimension of a length and will be called the range of thermal neutrons in carbon.

Quite similarly, since we assume that the uranium sphere is "black" for resonance neutrons, we can write for J^{res} the number of resonance neutrons absorbed by the sphere per second with good approximation

$$(9) \qquad J^{res} = 4\pi Q\,R B^2\left(1 + \frac{R}{B}\right)$$

where

$$(10) \qquad B = \frac{\lambda^{res}}{\sqrt{3}}\sqrt{k^{res}}$$

λ^{res} is the mean free path of resonance neutrons for scattering and B has the dimension of a length and will be called the range of the resonance neutrons in carbon.

Expression No. 9 is identical with the expression No. 8 which holds for a sphere which is "black" for thermal neutrons. Only the values for λ , the mean free path for scattering, and k , the average number of collisions which a neutron survives within the category called „thermal" or „resonance," are different for these two categories of neutrons. It would be strictly true that J the number of neutrons belonging to a category which will reach the sphere by diffusion per second is determined in the same way for different categories by λ , the mean free path, and $g(h)$, the function giving the probability of surviving h collisions with carbon atoms. In reality the function $g(h)$ is different for thermal and for resonance neutrons and expression No. 9 holds in so far as we may assume that J is determined with sufficient accuracy by λ and $k = \int_0^\infty h g(h)\, dh$ the first moment of $g(h)$

From No. 8 and No. 9 we find as the value of ε for a sphere which is "black" for thermal neutrons $\varepsilon = \varepsilon_0$

(11)
$$\varepsilon_0 = \frac{A^2}{B^2}\,\frac{1 + R/A}{1 + R/B}$$

Assuming, for example, $\sigma_{sc} = 4.8$; $\sigma_c = 0.005$ and $\dfrac{\lambda^{res}}{\lambda} = 1.18$

we find for graphite of density 1.7:
$$A = 43.5 \; ; \; B = 6.5 \text{ cm}$$

A real sphere of uranium having a finite radius is not "black" for thermal neutrons and J^{th}, the number of thermal neutrons absorbed by the sphere per second is smaller than J_0^{th} . We write

(12)
$$J^{th} = J_0^{th}\, \varphi$$

and accordingly we have

(13)
$$\varepsilon = \varepsilon_0\, \varphi$$

(14)
$$\varepsilon = \frac{A^2}{B^2}\,\frac{1 + R/A}{1 + R/B}\, \varphi$$

In order to calculate φ we take into account that inside the uranium sphere the thermal neutron density ρ obeys the equation

(15)
$$D(u)\frac{d^2(r\rho)}{dr^2} - S(u)\, r\rho = 0$$
$$D(u) = \frac{v\,\lambda(u)}{3}\,; \quad S(u) = v\, N_u\, \sigma_a(u)$$

having as its solution

(16)
$$\rho(r) = \frac{C}{r}\left(e^{r/u} - e^{-r/u}\right)\,; \quad r < R$$

where

(17)
$$u = \sqrt{\frac{D(u)}{S(u)}} = \sqrt{\frac{\lambda(u)}{3\, N_u\, \sigma_a(u)}}$$

and for pure uranium metal we have

(18)
$$u = \lambda(u)\sqrt{\frac{\sigma_{sc}(u)}{3\,\sigma_a(u)}}$$

From equations 3, 4, and 16 we find that g^{th} the number of thermal neutrons diffusing into the sphere per second is given by

$$g^{th} = g_0^{th}\,\varphi$$

where

(19)
$$\varphi = \frac{\dfrac{\lambda(u)}{u}\left\{\dfrac{e^{R/u} - e^{-R/u}}{e^{R/u} + e^{-R/u}} - \dfrac{u}{R}\right\}}{\dfrac{\lambda}{R}\left(1 + \dfrac{R}{A}\right) + \dfrac{\lambda(u)}{u}\left\{\dfrac{e^{R/u} - e^{-R/u}}{e^{R/u} + e^{-R/u}} - \dfrac{u}{R}\right\}}$$

For uranium in its pure state we have from No. 14, 18, and 19

(20a)
$$\varepsilon = \frac{A^2}{B^2}\times\left\{\frac{1}{1 + R/B}\times\frac{1}{\dfrac{\lambda}{RG\sqrt{\dfrac{3\sigma_a(u)}{\sigma_{sc}(u)}} - \lambda(u)}} + \frac{1}{1 + R/A}\right\}$$

Where G stands for

$$G = \frac{e^{R/u} - e^{-R/u}}{e^{R/u} + e^{R/u}}\,; \quad \frac{R}{u} = \frac{R}{\lambda(u)}\sqrt{\frac{3\,\sigma_a(u)}{\sigma_{sc}(u)}}$$

For $\frac{R}{u} > 2$ we can write $G \cong 1$, the difference being about 3.5% for $\frac{R}{u} = 2$.

The first factor in expression No. 20a increases proportionately with the reciprocal value of the capture cross-section of carbon. — The second factor is practically independent of the carbon cross-section since we have $\frac{R}{A} \ll 1$. Its value is determined by the density of graphite and uranium and the nuclear values $\sigma_{sc}(u)\,; \frac{\sigma_a(u)}{\sigma_{sc}(u)}$. The value of R may be chosen as to make this factor a maximum.[23]

All expressions for ε were so far obtained from diffusion equations involving the assumptions : $R \gg \lambda\,; \; R \gg \lambda(u)\,; \; u \gg \lambda(u)$. —

LATTICE OF URANIUM SPHERES

We shall now consider an infinitely large number of uranium spheres which may form, for instance, a close-packed lattice embedded in an infinite mass of carbon, and we shall determine q the ratio of the number of the thermal neutrons absorbed by the uranium spheres to the number of resonance neutrons produced in the system. We assume that the production of resonance neutrons is homogeneous throughout the whole mass of the carbon. The number of thermal neutrons produced in the carbon will be smaller than the number of resonance neutrons produced because a fraction of the latter is absorbed by the uranium spheres.

In the absence of the uranium spheres in the carbon the thermal neutron density ρ would have everywhere the same value ρ_0. In the presence of uranium spheres in the carbon the average neutron density $\bar{\rho}$ in the carbon is reduced by some factor α

$$\bar{\rho} = \alpha \, \rho_0$$

This reduction of the average thermal neutron density in the carbon by the uranium spheres is partly due to the absorption of resonance neutrons by the uranium spheres and partly to the absorption of the thermal neutrons by the uranium spheres. Since the number of neutrons captured per second by carbon is proportionate to the average neutron density $\bar{\rho}$ and since in the absence of uranium all the neutrons produced are captured by carbon, the fraction of the neutrons which is captured by carbon in the presence of the uranium lattice is given by α. Correspondingly, the fraction of the neutrons which are absorbed by the uranium lattice is given by $1-\alpha$. We can therefore write for q

$$q = (1 - \alpha) \, \frac{J^{th}}{J^{th} + J^{res}}$$

where J^{th} and J^{res} are the number of thermal neutrons and the number of resonance neutrons absorbed per second by one uranium sphere in the lattice.

We shall now calculate the values of J^{μ} and J^{res} from the values of Jth and J^{res} which were previously defined for a single uranium sphere in carbon. In doing so we shall neglect for the moment the fact that the production of the thermal neutrons in the carbon which contains a lattice of uranium spheres is not homogeneous, being smaller in the proximity of the uranium spheres than elsewhere. This effect, which is due to the absorption of resonance neutrons in the uranium spheres, will be taken into account later and will lead to a small correction which has to be applied to the final result.

If the distance L of the neighboring uranium spheres in the lattice is 10 large compared to the radius of the spheres the effect of the thermal neutron absorption of <u>one</u> uranium sphere on the thermal neutron absorption of its neighbors will be negligible. Nevertheless, the average thermal neutron density may be greatly reduced in the carbon by the presence of the uranium spheres, in particular if the range A of the thermal neutrons in carbon is large compared to the distance L. Under such conditions the average thermal neutron density determines with good approximation the number of thermal neutrons absorbed by one sphere in the lattice and we can write

(20)
$$g^{th} = \alpha \mathcal{I}^{th}$$

In reality, the thermal neutron absorption will be somewhat higher so that No. 20 represents a conservative value, but the correction is small if the volume of the uranium spheres is small compared to the volume of the carbon. This can be seen, for instance, from Equation No. 5 which shows that for large values of r/R the thermal neutron density is close to $\int_\infty$, even for a uranium sphere which is black for thermal neutrons.

Further, since the distance L between neighboring uranium spheres within the lattice will be large compared to B, the range of the resonance neutrons in carbon, and we have for g^{res}, the number of resonance neutrons absorbed by a uranium sphere within the lattice,

(21)
$$g^{res} = \mathcal{I}^{res}$$

From this it follows that q the fraction of all the neutrons which are absorbed by the uranium spheres in the thermal region ~~alone~~ is given by

(22)
$$q = \frac{\mathcal{I}^{th} \cdot \alpha}{\mathcal{I}^{th}\alpha + \mathcal{I}^{res}} (1 - \alpha)$$

or

(23)
$$q = \frac{\varepsilon \alpha}{1 + \varepsilon \alpha} (1 - \alpha)$$

This expression has its maximum value for $\alpha = \alpha_m$. The value of α depends on the spacing of the uranium spheres i.e. ~~on the~~ it depends on the mixture used and is ~~not~~ a property of the single uranium sphere

(24)
$$\alpha_m = \frac{-1 + \sqrt{1 + \varepsilon}}{\varepsilon}$$

and for q_m the maximum value for q we have (25)

(25)
$$\alpha_m = \frac{1 - q_m}{2}$$

(26)
$$q_m = 1 - 2 \frac{-1 + \sqrt{1 + \varepsilon}}{\varepsilon}$$

or

(27)
$$\varepsilon = \frac{4 q_m}{(1 - q_m)^2}$$

By calculating the value of q_m from ε we have neglected the effect of the absorption of resonance neutrons (by the uranium spheres in the lattice) on the production on thermal neutrons in the carbon in the neighborhood of the uranium spheres. The absorption of resonance neutrons reduces in reality the value of Q near the spheres below the average value of Q and accordingly the correct number g^{th}_{corr} of thermal neutrons absorbed by the uranium spheres per sphere will be smaller than g^{th}. Corresponding to this, the correct value, q_{corr}, of q, ~~will~~ will be smaller than q_m.

If the uranium sphere in the lattice did not absorb resonance neutrons then a neutron which reaches a given sphere at least once while its energy is in the resonance region would have a certain probability γ of reaching the sphere at least once after it had been slowed down to the thermal region. It can be shown that we have

$$q_{corr} = q_m \frac{1 - \frac{\nu\varphi(1-q_m)}{2q_m}}{1 - \frac{\nu\varphi(1-q_m)}{1+q_m}}$$

For values of $\nu\varphi < 0.667$; $q_m > 0.68$ we find $q_{corr}/q_m > 0.98$

In order to take into account this and other corrections and in order to deviate on the conservative side we shall in discussing the chances of a chain reaction use a value of q which is ten per cent below the calculated value

SPACING OF THE LATTICE

In deriving formula 26, it was assumed that throughout the whole mass of carbon in which the lattice of uranium spheres is embedded, there is a uniform production of resonance neutrons. We have to verify this assumption. Let $\sqrt{\bar{r}^2}$ denote the distance to which a fast neutron diffuses away from its points of origin before it is slowed down to thermal energies. Further, let L denote the distance between two neighboring uranium spheres in a close packed hexagonal cubic lattice. Then obviously, equation 26 was based on the assumption that the value of L, corresponding to the maximum value of q is not large, compared to the $\sqrt{\bar{r}^2}$. If it were otherwise, equation 26 ~~should~~ should not have been used to calculate the value of q_m.

In order to estimate L, as well as for other reasons, we shall calculate the volume V of carbon per uranium sphere in the lattice.

In the lattice of uranium spheres from the $Q V$ neutrons which are slowed down per second within the volume V to resonance energies, the carbon absorbs $\alpha Q V$ neutrons and the uranium spheres in the lattice absorb $(1-\alpha) Q V$

neutrons. Accordingly we have

$$g^{th} + g^{res} = (1 - \alpha) Q V$$

and from this we find

(31)
$$V = 4\pi \varphi \frac{\alpha}{q} A^2 R (1 + R/A)$$

and if q has its maximum value q_m we have

$$\frac{\alpha}{q} = \frac{1 - q_m}{2 q_m}$$

and we have

(32)
$$V = 4\pi \varphi \frac{1 - q_m}{2 q_m} A^2 R (1 + R/A)$$

This gives for the ratio of the volumes of carbon and uranium

(33)
$$\frac{V}{\frac{4\pi}{3} R^3} = 3 \varphi \frac{1 - q_m}{2 q_m} \frac{A^2}{R^2} (1 + R/A)$$

or using (14) and (27) we obtain

(33a)
$$\frac{\frac{4\pi}{3} R^3}{V} = \frac{1 - q_m}{6} \frac{R^2}{B^2} \frac{1}{1 + R/B}$$

For L the distance between neighboring uranium spheres in a hexagonal or cubic close-packed lattice we have

(34)
$$L^3 = \left(V + \frac{4\pi}{3} R^3 \right) \sqrt{2}$$

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BALANCE OF EMISSION AND ABSORPTION OF NEUTRONS BY URANIUM

A direct comparison of the number of fast neutrons emitted by uranium with the number of thermal neutrons absorbed by uranium may be obtained from different types of experiments carried out by Halban, Joliot and Kowarski,[11] Anderson, Fermi and Szilard,[12] and Halban, Joliot, Kowarski and Perrin.[13] These experiments show that on the average more than one fast neutron is emitted by uranium for one thermal neutron absorbed by uranium.

Halban, Joliot and Kowarski reported[8] that 3.5 ± 0.7 fast neutrons are emitted per fission. Anderson, Fermi and Szilard reported[9] that about $\mu = 1.5$ fast neutrons are emitted on the average for one thermal neutron absorbed by uranium. Recently Halban, Joliot, Kowarski and Perrin[13] carried out an experiment of the following type: a sphere of radius r_0 is filled with a homogeneous mixture of uranium and water which contains n atoms of hydrogen per atom of uranium. This sphere is immersed in a large water tank and a photo neutron source is placed in the center of the sphere. The density ρ of the thermal neutrons is measured along a radius and the integrals of

$$I_{int} = \int_0^{r_0} r^2 \rho \, dr \quad ; \quad I_{ext} = \int_{r_0}^{\infty} r^2 \rho \, dr$$

are determined, giving a measure of the number of slow neutrons which are present within the mixture inside the sphere and in the water outside the sphere. In another experiment the integral

$$I_0 = \int_0^{\infty} r^2 \rho \, dr$$

is determined for the same neutron source in pure water.

From the values of these three integrals which these authors determined for n = 1, 2 and 3 and r_0 = 25 cm. they concluded that on the average eight neutrons are emitted from the uranium within the sphere for one photo neutron which is slowed down to thermal energies and causes a fission process in uranium. This was interpreted to show that a considerable number of secondary and tertiary neutrons were generated in this experiment.

Since the value of the fission cross-section is not well known and rather difficult to determine, it seems necessary to avoid expressing results in terms of this cross-section and to use the magnitude μ rather than the number of neutrons emitted per fission. An accurate knowledge of the value of μ is of greatest importance from the point

of view of estimating the chances and finding the optimum conditions for a chain reaction. In the following we shall therefore indicate in what way an accurate value for μ can be obtained, from an experiment of the type performed by Halban, Joliot, Kowarski and Perrin:

Let F_{int} denote ~~F_{int}~~ the average number of neutrons which reach the thermal region inside the sphere within the uranium water mixture for one neutron emitted by the source in the center of the sphere, ~~is given by~~ We have then:

$$F_{int} = \frac{L_{int}/I_o}{\tau_{int}/\tau_o}$$

where

3> $$\frac{\tau_{int}}{\tau_o} = \frac{n\,\sigma_c(H)}{s\{\sigma_a(U) + n\,\sigma_c(H)\}}$$

is the ratio of the life-time of the thermal neutrons in the mixture inside the sphere and in pure water. ~~xxxxidxxthxxxphxxx~~

Of the neutrons which are slowed down to the resonance region inside the sphere a fraction p is absorbed at resonance and a fraction $(1 - p)$ reaches the thermal region. F_{int}^{*} the number of the neutrons reaching the resonance region per second inside the sphere is therefore given by

$$(1-p)\,F_{int}^{*} = F_{int}$$

F_{ext}^{*} the number of neutrons reaching the resonance region outside the sphere in the pure water is the same as F_{ext} the number of neutrons reaching the thermal region outside the sphere in the pure water and we have, therefore

$$F_{ext}^{*} = F_{ext} = \frac{I_{ext}}{I_o}$$

P, the total number of fast neutrons emitted by the uranium in the mixture for one neutron emitted by the neutron source in the center of the sphere is equal to the total number of the resonance neutrons produced minus 1, the neutron emitted by the source in the center. Therefore, we have

$$P = F_{int}^{*} + F_{ext}^{*} - 1 = \frac{F_{int}}{1-p} + F_{ext} - 1$$

The total number of fast neutrons produced by the uranium divided by the number of thermal neutrons produced within the sphere in the mixture is

$$\frac{P}{F_{int}} = \frac{1}{1-p} + \frac{F_{ext} - 1}{F_{int}}$$

And finally since the uranium absorbs ~~of~~ *from* the neutrons which reach the thermal region within the sphere the fraction

$$\frac{\sigma_a(U)}{\sigma_a(U) + n\,\sigma_c(H)}$$

we have

$$\mu = \frac{P/F_{int}}{\left(\dfrac{\sigma_a(U)}{\sigma_a(U) + n\,\sigma_c(H)}\right)}$$

or

$$\mu = \frac{\sigma_a(U) + n\,\sigma_c(H)}{\sigma_a(U)}\left\{\frac{1}{1-p} + \frac{C_{int}}{C_0}\,\frac{I_{ext}/I_0 - 1}{I_{int}/I_0}\right\}$$

Introducing the value of $\dfrac{C_{int}}{C_0}$ from No. 37 we obtain

$$38\quad \mu = \frac{1}{1-p} + \frac{n\,\sigma_c(H)}{\sigma_a(U)}\left\{\frac{1}{1-p} - \frac{1}{3}\,\frac{1 - I_{ext}/I_0}{I_{int}/I_0}\right\}$$

From this equation we see that we can determine μ by determining the value of p.

A considerable fraction of the fast neutrons emitted by uranium may of course escape from within the sphere but the value of this fraction need not be determined for our purpose. On the other hand, the formula given for μ holds with good approximation only if the sphere is sufficiently large to permit us to neglect transition phenomena in the equilibrium between resonance neutrons and thermal neutrons near the surface of the sphere. Since the uranium inside the sphere absorbs resonance neutrons the density of the neutrons which have an energy below the upper end of the resonance region of uranium and above the thermal region is smaller∧ This has the consequence that neutrons of this category will diffuse from the water across the sphere into the inside of the sphere. If the radius of the sphere were not sufficiently large so that this phenomenon could not be neglected the expression $\dfrac{F_{int}}{1-p}$ would then give a too large value for the number of resonance neutrons produced inside the sphere. Correspondingly the expression No. 38 would give a too large value for μ. But with the proviso that a sufficiently large sphere is used we shall obtain an accurate value for μ if only the value of p is measured accurately.

∧near the surface inside the sphere than near the surface outside the sphere.

The value of p has so far not been measured for hydrogen concentrations as low as n = 3 but we can give a lower limit for p for this concentration as we are able to extrapolate by means of theoretical considerations from values of p which have been measured for higher hydrogene concentrations (*i.e.* ~~such as~~ n = 30.) *which lead us to the formula:*

~~In order to do this we shall make use~~ of formulae relating to homogeneous mixtures of hydrogen and ~~uranium~~ uranium which will be published shortly by G. Placzek. A general treatment of the process of the slowing down of neutrons in water has been given by E. Fermi. *in 1936* In a ~~uranium~~ uranium oxide - water mixture p, the fraction of the neutrons which is absorbed by uranium between the energies E_2 and E, is given by [*]

39)
$$\ln(1-p) = -\int_{E_1}^{E_2} \frac{f(E)}{E}\, dE \qquad ; \quad E > 1\ Volt$$

where

40
$$f(E) = \frac{1}{1 + \frac{nH}{g(E)}}$$

$g(E)$ is the ~~capture~~ cross- section of uranium for radiative capture and H is the scattering cross- section of hydrogen for the resonance neutrons of uranium. $(H \doteq 17 \times 10^{-24}\ cm^2)$ If the hydrogen concentration is high enough so that the energy region in which there is an appreciable absorption by uranium is small compared with the resonance energy E_0 one may write

41
$$\ln(1-p) = -\frac{1}{E_0}\int_{-\infty}^{\infty} f(E)\, dE$$

~~This gives~~ for a single absorption line of the form

42
$$g(E) = \frac{g_0}{1 + \left(\frac{E-E_0}{\Gamma}\right)^2} \qquad ; \quad g_0 = g(E_0)$$

~~giving [*]~~

43
$$\ln(1-p) = -\frac{\pi\Gamma}{E_0}\frac{1}{\sqrt{\frac{nH}{g_0}\left(1+\frac{nH}{g_0}\right)}}$$

For $n \lessgtr 30$ we have $\frac{nH}{g_0} \ll 1$ and No 43 gives [**]

44
$$\ln(1-p) = -\frac{\pi\Gamma}{E_0}\sqrt{\frac{g_0}{nH}}$$

← From this we see that for such hydrogen concentrations for which No 44 holds we have for two different hydrogen concentrations n and n_1

45
$$\ln(1-p) = \sqrt{\frac{n_1}{n}}\ \ln(1-p_1)$$

***

[*] Private communication by G. Placzek June, 1939

[**] ~~Private communication~~ independently by *G. Placzek and* E. Fermi June , 1939

This relationship No. 45 does no longer hold for hydrogen concentrations which are as low as n = 3. But we may write in the case of uranium

(46)
$$\ln(1-p) = \int_{0.2E_0}^{1.8E_0} \frac{f(E)}{E} dE$$

and it is then easy to see that for an absorption line of the form given by No. 42 which is symmetrical in (E - E$_0$) or even more so for a similar line which deviates from symmetry in the sense that the absorption is larger for smaller energies (as it is the case for a line which obeys the Breit-Wigner formula), we have

(47)
$$\ln(1-p) < \sqrt{\frac{n'}{n}} \ln(1-p_1) \quad \text{if} \quad n < n_1$$

The value of p has been measured by Halban, Joliot, Kowarski and Perrin for n = 30 and was found to be p = 0.2 ± 0.02; using equation No. 47 we find from this

$$p_3 > 0.5 \pm 0.04 \quad \text{for } n = 3$$

According to No. 38 μ increases with increasing values of p and in the circumstances we should obtain a conservative value for μ by using the value of p = 0.5.

Using this value and the values

$$\frac{I_{int}}{I_0} = 0.72 \quad ; \quad \frac{I_{ext}}{I_0} = 0.45$$

reported by Halban, Joliot, Kowarski and Perrin for a sphere of 25 cm. radius filled with a uranium oxide--water mixture for which they had n = 3 and s = 0.42 gm./c.c. we find from No. 38

(48)
$$\mu = 2 + 0.2 \frac{3\sigma_c(H)}{\sigma_a(u)}$$

We see that the value obtained for μ if calculated from this equation is scarcely affected by the wide limits of error of the present experimental values of $\sigma_a(u)$. By attributing the value of ¼ or 1/8 to $\frac{3\sigma_c(H)}{\sigma_a(u)}$ we obtain $\mu = 2.05$ or $\mu = 2.02$ respectively.

In these circumstances we shall use ~~for the present as a presumably conservative value and~~ as the best value at the present available: $\mu = 2$

$$\mu = 2$$

Conditions For A Chain Reaction

Since q denotes the fraction of fast neutrons emitted by uranium which are slowed down to the thermal region and are absorbed as thermal neutrons by uranium and since μ denotes the number of fast neutrons produced on the average by uranium for one thermal neutron absorbed by uranium, then obviously $\mu q > 1$

is the condition for the possibility of a chain reaction. If this condition is fulfilled then a divergent chain reaction can be maintained in a sufficiently large system from which only a small fraction of the neutrons emitted by the uranium within can escape across the boundary of the system without being absorbed within.

Accordingly, the condition for the possibility of a chain reaction in a system composed of a lattice of uranium spheres embedded in carbon $\mu q_{corr} > 1$

and using equation No. 30, we find $0.9 \, \mu \, q \, m > 1$

From which we find by using equation No. 27 as a sufficient condition for the possibility of a chain reaction

(51)
$$\varepsilon > \frac{4.44 \, \mu}{(\mu - 1.01)^2}$$

Using the value of $\mu = 2$ we have as a sufficient condition $\varepsilon > 11.3$

In order to see now whether a chain reaction is possible we have to calculate from our formulae the numerical value of ε. We shall do that in the following under the assumption that the energy liberated in the chain reaction will maintain the carbon at a temperature of about 900 C° and in order to be on the conservative side *27 we shall assume that the temperature of the uranium spheres in which most of the energy is liberated is, in spite of efficient cooling, about the same.

Since we have at room temperature $\sigma_c(C) < 0.01$ we shall have at 900 C°. a capture cross-section of carbon half of this value. The scattering cross-section of uranium for thermal neutrons we take to be $\sigma_{sc}(U) = 9$. Finally, at room

temperature we take $\frac{\sigma_a(U)}{\sigma_{sc}(U)} = 1/2$ and correspondingly we take at 900° C. $\frac{\sigma_a(U)}{\sigma_{sc}(U)} = 1/4$

For a density of graphite of 1.7 and a density of uranium of 15 we then obtain

from No. 14 for R = 8 cm [*28] : $\varepsilon = 14$ This being larger than the value re-

quired by No. 51 we can just about expect a divergent chain reaction to take place

in the system for $\sigma_c(C) = 0.01$. In reality the capture cross-section of carbon

is perhaps much smaller than the upper limit which has so far been established and

consequently there is hope that conditions for a chain reaction will be quite

favorable.

The amount of carbon and uranium required to reach the point of divergence at

which nuclear transmutation will proceed at a rate limited only by the necessity of

avoiding over-heating is essentially determined by the value of $(\mu q - 1)$ If we

take the density of graphite to be 1.7; the density of uranium metal to be 15, and

choose R = 8 cm. [*28] We then obtain for a capture cross-section of carbon at

room temperature of $\sigma_c(C) = 0.005$ the following set of values at 900° C:

A = 64 cm; $\frac{A^2}{B^2} \approx 90$; $q = 0.666$; $\varepsilon = 27$; $q_m = 0.681$;

$q_{corr} = 0.66$; and for $\mu = 2$; we obtain $\mu q - 1 = 0.32$.

CRITICAL DIMENSIONS

For a large sphere of graphite which contains a large number of small spheres of

uranium, the critical value l for the radius of the graphite sphere for which the

chain reaction becomes divergent may be calculated for various distributions of uranium

within the graphite sphere. The optimum distribution of uranium is not uniform within

the graphite sphere and will either decrease or increase with r according to whether

we want to have a minimum amount of uranium or a minimum value for 1. The treatment of

this question may as well be postponed until the value of the carbon cross-section is

known. It will then be possible to find the optimum distribution of uranium as a

function of the distance from the center of the graphite sphere and give a value
for 1. In the meantime, a very rough approximation may give an idea of the order of
magnitudes which are involved. In graphite of 1.7 density some average distance $\bar{r}$
to which a fast neutron emitted by uranium diffuses away from its point of origin until
it becomes a thermal neutron and reacts with uranium or carbon is about 50 cm. For

the critical radius 1 *17 and 20 we have

$$l \sim \sqrt{\frac{3(\bar{r})^2}{\mu q - 1}} = \sqrt{\frac{7500}{\mu q - 1}}$$

and for $\left(\mu q - 1\right) = \frac{1}{8}$ we get l = 250 cm. This corresponds to about 100 metric
tons of graphite. If the carbon capture cross-section is lower, then l will be smaller.

The amount of uranium required can be calculated from equation No. 33a

$$\frac{4\pi}{3} R^3 / \nu = \frac{1 - q m}{6} \frac{R^2}{B^2} \frac{1}{1 + R/B}$$

It may be kept down by choosing a smaller value for R than the value corresponding
to the maximum value of Σ. For R = 5 cm. and q_m = 0.6 33a gives $\frac{4\pi}{3} R^3 / \nu = 0.0336$

corresponding to about 30 tons of uranium for 100 tons of graphite. *29

MEASUREMENTS

In order to have an accurate value for μ it will be necessary to have a direct
measurement *30 of p for small hydrogen concentrations preferably for n = 3.

The upper limit which has been reported for the carbon cross-section by Halban,
Frisch, and Koch is already so low that it would be difficult to improve upon it un-
less a method were used which is specifically designed to measure extremely small
capture cross-sections. Such a method will be described in the following:

Let us consider a sphere of carbon and a neutron source in the center of the
sphere. The thermal neutron density inside the carbon will then obey equation No. 3
only in this case Q is a function of r of which we must not assume $\frac{dQ}{dr} = 0$
Let the carbon sphere be immersed in a water tank or surrounded by paraffin wax. The
thermal neutron density will then have a certain fairly high value at the surface of
the sphere and inside the sphere it will be some function of r, $f_1(r)$. If in a second
experiment the surface of the sphere in the water is covered by cadmium and the thermal
neutron density at the surface of the sphere is then reduced to zero and inside the
sphere it will be another function of r, $f_2(r)$. The difference $f(r) = f_1 - f_2$
obeys the homogeneous equation

$$D(c) \frac{d^2(r f)}{dr^2} - \delta(c) r f = 0$$

which has the solution

$$f(r) = c \frac{e^{r/A} - e^{-r/A}}{r}$$

We can thus find A by determining the value of f for two values of r, for instance,

r = 0 and r = r. It is

$$\frac{\rho(r)}{\rho(0)} = \frac{e^{r/A} - e^{-r/A}}{2\,r/A}$$

or for small values of r/A

$$\frac{\rho(r)}{\rho(0)} \cong 1 + \frac{1}{6}\left(\frac{r}{A}\right)^2$$

Using a sphere of graphite of fifty to seventy centimeters of radius it should be possible to measure the range A with sufficient accuracy.

It is, however, quite easy artificially to stabilize the chain reaction by slowly shifting the position of absorbing bodies within the system in such a way as to reduce the average value of q whenever the intensity of the neutron radiation emanating from the chain reaction increases. One might perhaps think that the time within which such controlling action would have to take place is very short. We shall therefore now show that this is not so.

Of the neutrons which are emitted in the chain reaction by uranium only a fraction ψ is absorbed within the system and $1-\psi$ escapes across the boundary of the system without reacting with uranium. A stationary state can be maintained as long as

$$\mu q \psi < 1$$

We write

$$q \psi = f(T, x(t))$$

In order to indicate that this product is a function of the temperature T and also depends on a parameter such as the position of some absorbing or scattering body near or within the system which can be shifted by some controlling mechanism and thus be made a function of time t.

In order to have a large neutron production we must maintain a chain reaction near the point

$$\mu f_0 = 1$$

If this product becomes larger than one, as it may well happen then there is an exponential rise in the neutron production and accordingly also in the temperature. In case of a sudden small deviation from one

$$f = (1 + \xi) f_0$$

the time t_2 in which the number of neutrons doubles is given by

$$t_2 \cong \frac{1}{2\xi} t_1$$

where t_1 is the time which a fast neutron emitted by a uranium atom in the system would require to produce two fast neutrons if it is slowed down and absorbed within the system.

For instance, if we had a sudden change in f of 0.1%, as we well may have, and if we have $t_1 = 8 \times 10^{-3} sec$ then it would take $t_2 = 4$ seconds for the neutron production to double its value and accordingly there would be only an insignificant rise in the temperature if the control responded within 4 seconds.

It is easy to see that for a lattice of uranium spheres in carbon

the mean life-time of a thermal neutron within the system is given by

$$\tau_1 = \alpha \, \frac{\sigma_{sc}}{\sigma_c} \, \frac{\lambda}{v}$$

For $\sigma_c = 0.005$ at room temperature, for instance, we have for

$$q_m = 0.66 \quad \text{and} \quad \alpha_m = \frac{1-q_m}{2} = 0.17$$
$$\tau_1 = 2 \times 10^{-3} \, sec$$
$$\tau_1 \sim 4\,\tau_1 \sim 8 \times 10^{-3} \, sec$$

In calculating $t_2 = 4$ sec we did not take into consideration the fact that a fraction of the neutrons is emitted by uranium with a time delay of about ten seconds. Though this fraction is small it has a marked effect in leading to still longer times than those which we have estimated. But as for all practical purposes the time t_2 which we have found is already long enough we need not include for the present the delayed neutron emission in the treatment of the subject.

Figure 1 illustrates one way in which the chain reaction could be stabilized. It represents a uranium sphere which is surrounded by a spherical layer of liquid bismuth. Bismuth serves the purpose of cooling the uranium and could be used for this purpose without appreciably reducing the nuclear efficiency of the arrangement, that is, without appreciably diminishing the value of q. The short rod or disc composed of an element which strongly absorbs thermal neutrons, is near the center of the uranium sphere and is shielded by the uranium from thermal neutrons. This rod ~~or disc~~ can move within a tube ~~or slit~~ and its position may be controlled by the intensity of ~~xxxx~~ the neutron radiation emitted by the chain reaction. If the intensity of this radiation increases, the rod ~~or disc~~ may be automatically moved away from the center of the uranium sphere and ultimately, if required, entirely out of the interior of the uranium sphere. It would then absorb a larger and larger fraction of thermal neutrons, thereby reducing the value of q and would thus stabilize the chain reaction.

Conclusions

We conclude that we can expect a chain reaction to take place in a sufficiently large mass of graphite which contains, for instance, a close-packed hexagonal or cubic lattice of uranium spheres. The capture cross-section of carbon is likely to be smaller than the upper limit so far established and consequently there is hope that moderately large masses of graphite and uranium or uranium oxide will be sufficient to reach the point of divergence at which nuclear transmutation can be maintained at an intensity which is limited only by the necessity of avoiding overheating.

Large quantities of radio-active elements will be produced directly from the splitting uranium atoms and indirectly by the intense neutron emission. The necessity of protecting human beings from irradiations emanating from the chain reaction may slow down the industrial development of this field, but it is difficult to imagine that practical applications should not follow in due course of time.

QUOTATIONS

0) H. G. Wells, The World Set Free, (1913)

1) Anderson, Booth, Dunning, Fermi, Glasoe and Slack, Phys. Rev. 55, 511, (1939)

2) Roberts, Meyer and Wang, Phys. Rev. 55, 510 (1939)

3) Halban, Joliot and Kowarski, Nature 143, 479, (1939)

4) Anderson, Fermi and Hanstein, Phys. Rev. 55, 797 (1939)

5) Szilard and Zinn, Phys. Rev. 55, 799, (1939)

6) v. Droste, Naturwissenschaften, 27, 198, (1939)

7) Haeny and Rosenberg, C. R. 208, 898, (1939)

8) Michiels, Perry, and G. P. Thomson, Nature, 143, 760, (1939)

9) v. Droste and Reddemann, Naturwissenschaften, 27, 371, (1939)

10) Rotblatt, Nature, 143, 852, (1939)

11) Halban, Joliot and Kowarski, Nature 143, 680, (1939)

12) Anderson, Fermi, and Szilard, Phys. Rev. 56, 284, (1939)

13) Halban, Joliot, Kowarski and Perrin, Journ. de Phys. 10, pp. 428-429, (1939)

14) F. Perrin, C. R., 208, 1394, (1939)

15) Adler and Halban, Nature 143, 793, (1939)

(16) G. Placzek. Phys. Rev. 55, 1130, 1939

(17) Francis Perrin. C. R. 1939.