NUCLEAR FISSION AND ATOMIC ENERGY

by

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by

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Chapter 11

FAST NEUTRON CHAIN REACTION

11.1 Possibility of a Fast Neutron Chain Reaction, Mathematical Theory

The use of a fast neutron chain reaction to produce large neutron pulses and to release large amounts of energy in a short time has many interesting applications. The feasibility of such reactions can best be studied by considering the diffusion equation first discussed in section 9.3. While the mean free path of the neutrons is comparable with the dimensions of fissionable material used and thus limits the validity of the calculation, nevertheless the analysis will show what quantities are important and will suggest their orders of magnitude.

The neutron density at any point will vary with time as a result of diffusion of the neutrons, and also because of absorption of neutrons by capture, and the production of new neutrons by fission. The appropriate differential equation for describing this variation is

$$\frac{\partial N}{\partial t} = D\nabla^2 N + KC \qquad (11.1)$$

These quantities have all been previously defined in section 9.3 but it is worthwhile to recall their dependence on other more physical quantities. The diffusion coefficient is

$$D = \frac{\lambda v}{3} \qquad (11.2)$$

where v is the velocity of the neutrons. The mean free path, λ, of the neutrons is given by

$$\lambda = \frac{1}{\sigma_s N_u + (\text{other scattering processes})} \qquad (11.3)$$

where σ_s is the cross section for scattering and N_u the concentration of the main constituent of the reacting material.

$$K = v\,\sigma_f\,N_U\,(\mu - 1) - (\text{other processes which use up neutrons}) \tag{11.4}$$

where σ_f is the cross section for fission and μ is the number of new neutrons released per fission.

We will assume a sphere of radius R and investigate the solution subject to the following boundary conditions:

$$N\,(r,0) = N_o,$$

$$D\,\frac{\partial N}{\partial r} + a\,N = 0,$$

at $r = R$. This second equation provides for continuity in the neutron current at the surface of the sphere. <u>a</u> is related to the rate at which neutrons leave the surface per unit area per unit concentration.

Conveniently, the solution of this problem is given in Byerly ()

() Byerly, Fourier's Series and Spherical Harmonics, (Ginn and 'Co., 1895) pp 117-122

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and may be written as

$$N\,(r,t) = \frac{1}{r} \sum_i A_i \exp\left[\left(K - \frac{Dx_i^2}{R^2}\right) t\right] \sin\frac{x_i r}{R} \tag{11.5}$$

where the x_i is the i'th root of the equation

$$\tan x = \frac{x}{1 - \frac{aR}{D}} \tag{11.6}$$

If the concentration of neutrons is to increase with time, the coefficient of the time in one of the exponentials must be positive. Thus, if x_0 is the smallest root of (11.6), a chain reaction will proceed when

$$R_C \geqq x_0 \sqrt{\frac{D}{K}} \tag{11.7}$$

Before we can determine κ_0 we must find the value of a. Solving equation 10.46 for $\left\{-D \frac{\partial N}{\partial r} / N\right\}_r = R$

$$a = \frac{1}{2} \; v \, \frac{(1-p)}{(1+p)}$$

where p is the fraction of neutrons reflected back.

From the previous definitions

$$\tan x_0 = \frac{x_0}{\left(1 - \frac{a}{\sqrt{DK}} x_0\right)}$$

$$\frac{a}{\sqrt{DK}} = \frac{(1-p)/(1+p)}{\sqrt{\frac{4}{3}(\mu-1)\,\sigma_f/\sigma_s}} \tag{11.8}$$

$$\sqrt{\frac{K}{D}} = N_U \sqrt{3\,(\mu-1)\,\sigma_f\,\sigma_s} \tag{11.9}$$

We have assumed that fission is the only important process and have neglected the other absorbing processes mentioned in (11.3) and (11.4). Further we have assumed in this discussion that the velocity of all neutrons is the same and that the cross sections are independent of neutron velocity.

11.2 Possibility of a Fast Neutron Chain Reaction, Calculations for U 235

Now let us examine the expression for the critical radius expressed in terms of measurable physical constants.

$$R_c = \frac{x_o}{\left(N_U \left\{3(\mu - 1)\,\sigma_f\,\sigma_s\right\}^{\frac{1}{2}}\right.} \qquad (11.10)$$

Obviously, we want x_o to be as small as possible and all the other quantities to be as large as possible. The value of x_o can be decreased by the use of a tamper which will reflect as many neutrons as possible. To increase N_u the material should be pure uranium 235. μ is taken as 2.3 and assumed to be the same for all fissionable material. The value of σ_s for fast neutrons probably does not vary as much for various nucleii as does σ_f. However, when σ_f is large, σ_s will also be large for the following reason. As far as the diffusion equation is concerned σ_s is the sum of all processes which scatter neutrons including fission, In the latter case, although one neutron disappears and μ new neutrons are actually produced, we regard it as consisting of the scattering of one neutron plus the production of $\mu - 1$ new ones.

The ideal material for producing a chain reaction is one in which every neutron entering the nucleus will produce a fission. The existence of competitive processes such as absorption and gamma ray emission decrease both σ_f and σ_s. An additional contribution to the "scattering" cross section is inelastic scattering in which the neutron is absorbed and another neutron reemitted with a lower energy. As far as calculation of the critical size is concerned, it is not necessary to know the relative contribution of the various scattering processes. Measurements of σ_s for non-fissioning heavy nuclei have been made by Dunning et al (). The fact that

() Dunning, Pegram, Fink and Mitchell, Phys. Rev. 48, 265 (1935)

roemission of a neutron after capture is far more likely than a radiative transition ( ) simplified their measurements. σ_s showed a regular in-

( ) Bethe, Rev. Mod. Phys. 9, 160 (1937)

crease proportional to $A^{2/3}$ and would be about 6×10^{-24} for uranium and plutonium. In U^{235} fission replaces neutron emission as the main process but, as mentioned above, this does not change the effective σ_s.

The average energy of the neutrons being considered is of the order of 1 Mev and for them σ_f is not simply the πR^2 mentioned in section 7.3 because their effective wave lengths are of the order $\lambda \sim 10^{-12}$ cm which is comparable with the nuclear radius R as determined by α particle scattering. Rabi () has shown that the measured cross section may be several times larger than πR^2, which fact accounts for the large value

() I. I. Rabi, Phys. Rev. 43, 858 (1933)

of σ_s.

The exact value of σ_f depends on the neutron energy and we can estimate an approximate value of, say, 3×10^{-24} cm, this being about half the total scattering cross section for U^{235}. A survey of the published cross sections of heavy non-fissioning nucleii indicates that potential scattering accounts for about half the total scattering.

The other half of the fast neutron scattering cross section is contributed by inelastic cross section effects, the most likely one being fission in those nuclei which will fission with thermal neutrons. This was discussed in section 8.6.

Inaccuracies in the values of σ_f and σ_s change our estimate of the critical mass seriously because of the rapid change in mass with radius. Smyth's description (12.32) of the extensive fast neutron cross section measurements at Los Alamos emphasize the importance of these cross sections. The calculation presented here should be considered as tentative in as much as in a report to the National Academy in November 1941 (Smyth 4.99) the critical mass could only be fixed as between 2 and 220 lbs. mainly on account of the uncertainties in the cross sections.

In the following calculations μ is taken as 2.3 neutrons per fission and

$$N_U = \rho \frac{Na}{M} = \frac{19 \text{ gm cm}^{-3} \times 6 \times 10^{23} \text{ atom (gm mol)}^{-1}}{235 \text{ gm (gm mol)}^{-1}}$$

$$= 4.8 \times 10^{22} \text{ gm cm}^{-3}$$

If all the neutrons reaching the surface escape, the reflection coefficient is zero and x_o is 2.10. It should be noted that in Adler's () calculation

() M. F. Adler, Comptes Rendus 209, 301 (1939)

he assumed that the concentration at the surface of the sphere was zero and for this case the value of x_o is π. This assumption is good in systems which are so large that $\lambda \ll R$. In our system $\lambda \sim R$ so that we use the more exact calculation; this results in a reduction in the critical mass by a factor of 3.4.

The critical radius with no tamper is R_c = 5.15 cm or a mass of about 23 pounds of U^{235}. The mean free path is given by $\lambda = \frac{1}{\sigma_t N_U}$ and for U^{235}, it is 3.4 cm. This value is quite comparable with the critical size and hence our use of the diffusion theory might lead to considerable error. If we compare the solution for the one dimensional random walk problem ( ) with the diffusion equation solution it appears that the probability of escape is larger than that calculated for diffusion. This simple consideration indicates that replacing the diffusion theory calculation by a more accurate statistical treatment of the neutron paths would give a slightly larger estimate of the critical radius.

( ) S. Chandrasekhar, Rev. Mod. Phys. 15, 1 (1943)

11.3 Effect of a Tamper

By the use of a tamper the critical size may be somewhat reduced. A value for the reflection coefficient can be found by solving the differential equation for two concentric spherical media, the inner sphere of U^{235} with the values of K and D discussed above, the second sphere is the tamper with about the same value of D but with $K = 0$, and the region surrounding it consists of empty space. Such a detailed calculation is hardly worth while at this point so a value for the reflection coefficient will be es imated very crudely.

In our approximation the value of the reflection coefficient p for a tamper of given thickness cannot be calculated until the critical size is known. Consequently a preliminary value of p will be used to get a preliminary value of R_c. This value of R_c will be used to redetermine p and hence calculate a more accurate value of R_c. Since there are advantages (as we shall see) in using a tamper of high density the mean free path in the tamper will be about the same as that in the fissionable material, 3.4 cm.

We assume tentatively that this is of the same order as the radius of the fissionable material. Then consider neutrons leaving the core perpendicularly as shown in Figure 11.1. On the average these neutrons will suffer a collision at a distance from the surface of the sphere, whereupon they will be scattered in all directions. The mean probability of being scattered into the solid angle subtended by the core is $\frac{\omega}{4\pi}$, where $\frac{\omega}{4\pi} = \frac{1}{2}(1 - \cos\theta)$, and $(\sin\theta)_{ave} = \frac{R}{R + \lambda}$. Therefore, p is roughly given by

$$p = \frac{1 - \sqrt{1 - \frac{R^2}{(R + \lambda)^2}}}{2} \qquad (11.11)$$

This estimate neglects all multiple scattering processes. However these effects tend to compensate since the neutrons which eventually return to the core after several scatterings would increase the scattering coefficient while neutrons which are scattered into the solid angle ω may be scattered out of it before reentering the core, and this would decrease p. Of course, all the neutrons do not leave the surface of the core normally as assumed, but this complication does not change the order of magnitude of the reflection coefficient and it is neglected here.

For the preliminary critical radius R_c = 5.15 cm, the reflection coefficient p is 0.10. The new critical radius is R_c = 4.65 cm or a mass of 16 lbs. The constancy of the calculations could be improved by repeating the calculation for a new value of the reflection coefficient based on the new critical radius.

A more indirect method of estimating the critical size can be made by using remarks in the Smyth report (6.39). The theoretical studies of Manley, Oppenheimer, Serber and Teller indicated that the energy release in a fast neutron chain reaction could be made greater than that estimated in the third report of the National Academy. It was indicated there (Smyth (4.99)) that between 1 and 5 percent of the fission energy should be released at a fission explosion.

A war department release stated that a typical fission explosion contained the explosive equivalent of about 20,000 tons of T.N.T. The fission of 8 pounds of U^{235} will produce this amount of energy. If the chain reaction has an efficiency of between 10 and 30 percent the total mass of U^{235} lies between 24 and 80 pounds. It thus appears that substantially more than the critical amount of material was used in these explosions.

If the U^{235} is not pure, the critical radius will be somewhat larger in virtue of the decrease in K for two reasons: the fission cross section for U^{238} is about one fifth that of U^{235}, and neutron absorption will use up some of the neutrons. In fact, pure uranium metal will not produce a chain reaction, even for an infinite sphere (Smyth (12.10)). The diffusion coefficient, D, will vary little with concentration. The percentage increase in the critical radius is probably about half the percentage concentration of U^{238} in the U^{235}. In the same manner, the separation of Pu^{239} from U^{238} need not be taken to completion. No estimate of the critical size for Pu^{239} will be made except to indicate that the value of σ_s and σ_f are both probably a little larger than for U^{235} and hence the critical radius is somewhat smaller. The energy release per fission may also be a little larger since the electrostatic forces are increased.

We now make an estimate of the rate at which the neutron density will build up in a spherical mass of fissionable material which is larger than the critical size. From equation (11.5) the time necessary for the neutron density to increase by a factor $e = 2.718$ is

$$T = \frac{1}{K - \frac{Dx_o^2}{R^2}} = \frac{1}{\frac{Dx_o^2}{R^2}\left[\left(\frac{R}{R_c}\right)^2 - 1\right]} \qquad (11.12)$$

If we let $R = R_c + \Delta R$, with $\Delta R \ll R_c$, then

$$T = \frac{R_c}{2K\Delta R} = \frac{1.7 \times 10^{-9}\ R_c\ \text{sec}}{\Delta R\ (\text{cm})}, \qquad (11.13)$$

since 1 for 1 Mev neutrons in U^{235}, K is of the order of $3 \times 10^8\ \text{sec}^{-1}$.

11.4 Production of Controlled Neutron Pulses

For experimental purposes, if a large controlled pulse of neutrons is desired, it can be obtained by combining several parts, each of which is smaller than the critical size, and disassembling them in a time comparable with the time for the neutron density to double.

As the neutron density builds up it will develop a pressure tending to blow the material apart. This pressure at the surface of the sphere has two principal contributions, the gamma radiation and the neutrons. The range of the beta rays and fission products is so short that their kinetic energy is soon shared with other nuclei and the energy is propagated as a shock wave at a velocity small in comparison with that of the neutrons and gamma rays. We can make a crude estimate of the neutron pressure, P, in terms of a gas composed of the 1 Mev neutrons and having a density N_o.

The pressure is crudely given by

$$P = \frac{1}{3} \quad N_0E$$

where E is the neutron energy. This places an upper limit on the pressure, since the neutrons are not reflected at the boundary as in a gas but travel several cm into the tamper before undergoing a collision.

Let us suppose that the tamper will withstand a pressure wave of short duration of 10^4 atmospheres. Then we can tolerate a neutron density of 10^{16} per cm^3. If the original neutron density were of the order of 1 per cm^3, the density would have to increase by a factor of e^{37} in order to reach a density of 10^{16} per cm^3. The total time required is

$$T^* = \log_e 10^{16} \times 1.7 \times 10^{-9} \frac{R_c}{\Delta R} = 37 \times 1.7 \times 10^{-9} \frac{R_c}{\Delta R} \text{ s}$$

and if $\frac{R_c}{\Delta R}$ is of the order of 1000, the time becomes about 5×10^{-5} sec. If controlled neutron pulses are to be produced, a mechanical motion of ΔR must be achieved within this time or the reacting material could not be disassembled before an explosion took place. For a radius of 5 cm the velocity of the moving parts must be the order of 100 cm/sec if the critical radius is exceeded by 0.1%. The velocity required of the moving parts varies inversely as the square of the fraction by which the critical radius is exceeded.

As mechanisms with parts moving near the speed of sound are feasible, neutron pulses of several microseconds duration and with an intensity of 10^{24} neutrons per second appear possible. The existence of such pulses would allow the application of many of the timing techniques of the radar art to a large number of problems. However, the possibility of a mechanical failure while the material is over critical is not attractive.

A rough estimate of gamma radiation pressure can be made by assuming that a one Mev gamma ray is given off for every neutron. The momentum of the gamma ray is roughly 1/50 that of the neutron of the same energy so their contribution to the pressure is of secondary importance. The gamma radiation, however, reaches the tamper material first so it may be of importance in a detailed calculation.

In the discussions in this chapter it is assumed that the neutrons are emitted instantaneously during a fission. It is also possible to construct assemblies which are overcritical for delayed neutrons but undercritical for fast neutrons. Hence the time constant is considerably longer. Smyth (12.46) describes experiments of this general type which were performed at Los Alamos.

11.5 Production of Single Pulses with Maximum Number of Neutrons

If the material is over critical and is not intentionally disassembled, the reaction will continue until the material is consumed, the fission products "poison" the reaction or the material is blown apart. In contrast to piles, there is probably little "poisoning" by the fission products since their most likely reaction with fast neutrons is scattering rather than absorption. If the critical radius has been exceeded by an amount ΔR and we further assume that there is no motion of the material or capture of the neutrons by the fission products, then the reaction will continue until the concentration of U^{235} has decreased by roughly twice $\Delta R/R_c$. In most cases the kinetic energy of the fission products will disperse the material long before the material has been consumed.

The number of neutrons required to produce fission products of the same energy as 20,000 tons of TNT is of the order of 10^{25} neutrons or a density of the order of 10^{22} neutron per cm^3. The pressure developed by a neutron gas in which the average energy of the neutron is of the order of 1 Mev may be about 10^{10} atmospheres. In a gun barrel a pressure of 10^4 atmospheres is considered to be a limit of pressure without damage. If pressures of a billion atmospheres are to be developed, extremely short times for building up the neutron density are required in order that inertia can be used to effectively hold the material together. The time required for the neutron pressure to build up from 10^4 to 10^{10} atmospheres is $1.7 \times 10^{-9} \frac{R_C}{\Delta R} \times (2.3 \log 10^6)$ or about $2 \times 10^{-8} \frac{R_c}{\Delta R} \approx \frac{10^{-7}}{\Delta R}$. To get an idea of the order of magnitudes involved let us assume that the tamper is 4 cm thick and composed of a dense material. The time required to move it through a distance ΔR under the influence of a static pressure P is approximately

$$t = \sqrt{\frac{8 \Delta R \rho}{P}}$$

where ρ is the density of the tamper.

For a pressure of a billion atmospheres (calculated for U^{235}) this time is approximately $10^{-7} \sqrt{\Delta R}$. Thus if the time for the neutron density to build up is to be less than that required to overcome inertial forces,

$$\frac{10^{-7}}{\Delta R} \leq 10^{-7} \sqrt{\Delta R}$$

or ΔR is approximately 1 cm. In other words, the critical radius must be exceeded by 20%, and thus the total mass increased from 16 to 28 lbs.

11.6 Assembly of Reacting Material

We must now consider methods by which the fissioning material can be brought into this over critical state in a time less than that required for the neutron density to build up to a pressure of 10^4 atmospheres. In order to make a rough estimate of the times involved we assume that the method of assembly is such that the critical radius is instantaneously exceeded by 20%. The time required for the neutron density to increase from, say, 1 neutron cm^2 to 10^{16} neutrons cm^3 is by the application of equation 11.13, 2×10^{-7} sec. Actually we can tolerate a time of assembly somewhat larger than this for in the early stages when R is small, the neutron density builds up more slowly. As Smyth (12.20) suggests there may be some advantage in introducing some material which will absorb neutrons during the early stages of assembly but which will be consumed by the time the critical radius has been exceeded by the maximum amount. In any event it appears that the maximum length of time allowed for over critical assembly must be made of the order of ten microseconds. To move a distance of 1 cm in this time will require a velocity of 10^5 cm per second and this is about the maximum velocity which can be achieved using standard ordnance practice.

There is another principle by which the building up of the neutron density during assembly can be delayed. The neutron density can be kept much lower than the initial value of 1 neutron/cm^3 which was assumed in the previous paragraph. The limit is set by cosmic rays which contribute about one thermal neutron per cm^2 per second or a density of $\frac{1}{2} \times 10^{-5}$ neutrons per cm^3. Let us introduce some Be covered by a thin (10^{-5} cm) layer of some material which will shield it from the alpha particles being emitted by the U^{235}. Then when the critical size is exceeded by nearly the maximum amount the shield can be rendered ineffective and the neutrons emitted by the Be (α, n) reaction will trigger the chain reaction.

In our discussion so far we have assumed that the method of assembly was by combining two or more pieces each of which is smaller than the critical size. Methods in which the material is rendered over critical by the removal of some neutron shield are not attractive inasmuch as no materials having extremely large cross section for fast neutrons are available, and secondly the times required for moving an object initially at rest through a fixed distance are much larger than that required for an object which has a large relative velocity. A neutron shield can be most easily constructed from a material of low atomic number which will quickly reduce the energy of the neutrons in the elastic collisions and hence decrease their ability to produce fission or render them easily captured by an added impurity.

There is a third method of bringing the system into the overcritical condition in which the tamper and U^{235} core of fixed mass are compressed so as to increase its density. We will show that the critical radius decreases much more rapidly than the actual radius so that such a compression, starting with a lump that is slightly under critical size, can easily reach the critical condition. This is illustrated in Fig. 11.2. The critical radius varies with the density of the U^{235} as

$$R_c \sim \frac{1}{C_U} = \frac{1}{\rho}$$

For a given mass the radius vs density is $R \sim \frac{1}{\sqrt{\rho}}$. Thus, starting at the point A we travel along the dotted compression curve as compression builds up. At the point B the critical state has been reached. In order to obtain a large release of neutrons or energy it would be desirable to continue the compression to a point such as C where the critical radius has been exceeded by a significant amount.

Increasing the density of tamper will increase its reflecting power as can be shown from equation 11.11. Furthermore the shorter mean free path implies that the neutrons will return to the core in a shorter time and thus increase the efficiency of the explosion.

The material can be brought into the over critical condition by means of a spherically symmetric implosion, but the practicability depends on whether sufficiently high pressures can be developed rapidly enough (10^{-5} sec) to overcome the elastic forces and inertia of the core and tamper. A pressure of the order of 10^6 atmospheres is required. This is about two hundred times the pressure developed in the combustion of slow burning powder in large guns. By the use of fast burning power the required pressures can conceivably be developed. It is beyond the scope of this book to discuss these ordnance problems in more detail. The problem of developing a spherically symmetric implosion in an extremely short time is a formidable one. If during the implosion the core of the assembly is distorted very much from a sphere the efficiency of the reactions will be reduced and a self sustaining reaction might not even occur. It might be desirable to start with an elongated unit which will become spherical during an asymmetrical implosion.

If the elastic forces arising in the compression are large they can be reduced somewhat by putting a hole at the center of the core which can be filled as the implosion develops. Such a hole will also be effective in reducing the rate at which the neutron density builds up during the early stages.

There is the possibility of producing a more energetic reaction by making the core material spongy and therefore less dense. For example, if we halve the density the critical radius doubles and the amount of material increases by a factor of 4. If the efficiency of the reaction is unchanged the explosion should be four times more energetic.

11.7 Choice of Tamper Material

The most important characteristic of the tamper is that it have the highest possible density. The inertial effect is important in containing the explosion. As the fast neutron scattering cross sections of materials of large atomic number is about the same, the mean free path will be smallest in the material of highest density. We recall that a short mean free path increases the reflection by the tamper and also the neutron effectiveness by returning the neutrons quickly. For materials of the same density, the one having the larger compressibility is to be preferred for the implosion type of assembly. In this connection also the tamper should not be too thick for its inertial effects will increase the difficulty of making an effective implosion. In line with these considerations, the most likely tamper materials appear to be gold or ordinary uranium. Uranium has the advantage that it has about the same density, higher atomic number and larger compressibility than gold. Furthermore, any fissions which occur in the tamper will tend to increase the total energy released in the reaction.