

# THE PHYSICS OF GRAPHITE MODERATED REACTORS\*

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## INTRODUCTION

Since 1942 when the first graphite moderated reactor was built in Chicago by Fermi and his collaborators, reactors have attracted considerable attention. Apart from its use as a source of high neutron flux, a reactor is the only means known at present of producing controlled atomic energy. A great deal regarding the physics and kinetics of a reactor has been published, although the chemical and metallurgical aspects of the problem are still treated as a secret. In the present article—emphasizing mainly the physical aspects of the various processes involved—we give a brief account of the method by which the critical size of a reactor is calculated.‡ After a short discussion of neutron economy, reactor control and power, we describe as an example, the Oak Ridge National Laboratory Graphite reactor.

All reactors are classified broadly into three groups (i) fast, (ii) intermediate and (iii) thermal, according to the neutron energy which is responsible for the majority of fissions in uranium. We shall consider here thermal reactors only, because of their importance as power generators and also because lot more is known about them. In order to thermalize the fast neutrons that are produced during fission one generally uses as a "moderator" any one of the following: heavy water, graphite, light water or beryllium. Further one may use natural uranium (99.3 per cent of  $U^{238} + 0.7$  per cent of  $U^{235} +$  traces of  $U^{234}$ ) or uranium enriched in  $U^{235}$  isotope, as "fuel". The uranium moderator assembly may be homogeneous or heterogeneous. To make our problem more specific we will consider a graphite moderated, heterogeneous thermal reactor using natural uranium. It may be mentioned here that in a homogeneous mixture of graphite and natural uranium a chain reaction cannot be sustained without the help of an external source.

Let us consider, the neutron cycle in a lattice of graphite and uranium (Fig. 1).

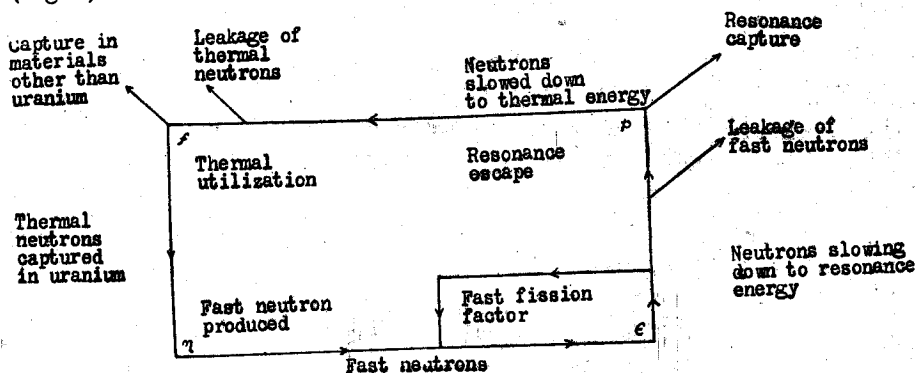


Fig. 1 Neutron Cycle in reactor. For infinite reactor there is no leakage of neutrons.

\* Based on a talk given by the author at the Defence Science Laboratory in Oct. 1954.

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‡ For details of calculation we should refer the reader to Glasstone and Edlund (1952) and Guggenheim and Pryce (1953). For details regarding reactor construction see Murry (1954) and Stephenson (1954). An elementary treatment of the reactor theory will be found in Soodak and Campbell (1954).

In order to avoid the complications introduced by the leakage of neutrons from a finite reactor we first treat an infinite lattice. The capture of one thermal neutron in natural uranium will lead, on the average, to the production of say,  $\eta$  fast neutrons (of energy  $\approx 2$  Mev). Some of these fast neutrons may lead to fission in  $U^{238}$  (fission threshold of  $U^{238}$  is nearly 1.0 Mev), so that number of neutrons slowing down past the fission threshold of  $U^{238}$  will be larger than  $\eta$ . If we define  $\epsilon$ , the fast fission factor, as the number of neutrons slowing down past the fission threshold of  $U^{238}$  per primary fission neutron, then the number of neutrons slowing down below 1.0 Mev per thermal neutron absorbed in uranium would be  $\eta\epsilon$ .

During further slowing down in the lattice some neutrons may suffer resonance capture in  $U^{238}$  (most important resonance of  $U^{238}$  is at 6.7 ev), and the number that escapes capture is determined by the resonance escape probability,  $p$ . These neutrons are finally thermalized. Of the  $\eta\epsilon p$  thermal neutrons (most probable energy 0.025 ev) some are captured in uranium and the rest in other materials. We define thermal utilization factor,  $f$  as

$$f = \frac{\text{number of thermal neutrons captured in uranium}}{\text{total number of thermal neutrons captured in the lattice.}}$$

From the above definition it would be clear that for an infinite lattice *i.e.*, when there is no leakage of neutrons, the reproduction factor  $k_{\infty}$ , which is defined as

$$k_{\infty} = \frac{\text{number of thermal neutrons in the present generation}}{\text{number of thermal neutrons in the preceding generation}}$$

is given by  $k_{\infty} = \eta\epsilon pf$ . This is known as the four factor formula. An infinite reactor would be critical, *i.e.* a self-sustained chain reaction would be maintained, if

$$k_{\infty} = 1 \quad \dots \dots \dots (1)$$

For a finite reactor the consideration of the leakage of neutrons is very important. In this case the criticality condition is written as

$$k = Pk_{\infty} = 1 \quad \dots \dots \dots (2)$$

where  $P$  is the non-leakage probability, *i.e.*, the probability that a neutron does not leak out of the lattice, and  $k$  is the effective reproduction factor. As  $P$  will always be less than unity, for a finite reactor the reproduction factor  $k_{\infty}$  has to be greater than unity. In order to calculate the critical size of a reactor one has to know the values of the four factors involved in  $k_{\infty}$  and also the value of  $P$ . In passing we may note that for a homogeneous reactor  $k_{\infty}$  depends only upon the nuclear constants of the materials comprising the reactor, while  $P$  in this case is determined by the size and shape of the lattice.

#### Determination of Reactor Constants

We now consider how the various factors that comprise  $k_{\infty}$  and  $P$  are determined for a heterogeneous thermal reactor using natural uranium. As we shall see some quantities are obtained experimentally whereas others have to be calculated.

(i)  $\eta$ : the average number of neutrons produced by the absorption of a thermal neutron in uranium has to be determined experimentally. The normally expected value is  $\eta = 1.308$ .

(ii) *Fast fission factor.* We assume the reactor to be cylindrical with uranium rods arranged in a square lattice parallel to the axis of the cylinder.

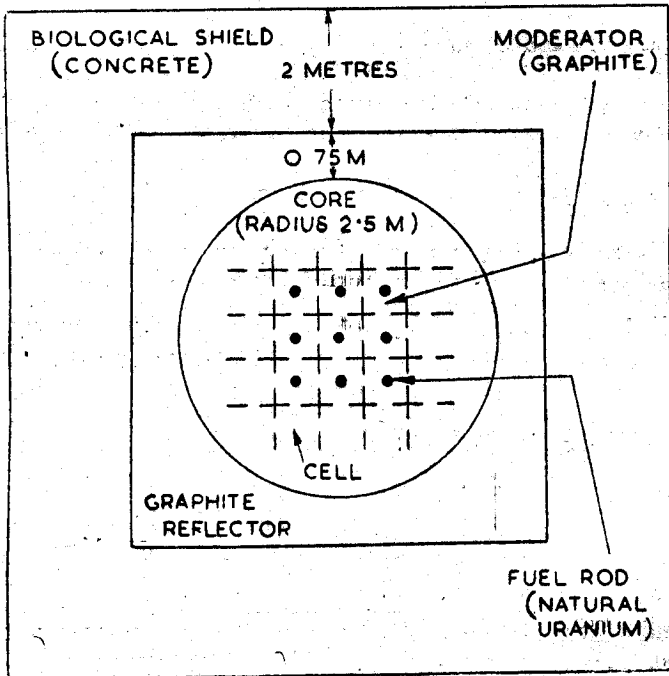


Fig. 2. Cross-Section perpendicular to the axis of hypothetical, graphite moderated, natural uranium reactor.

For the sake of calculations one replaces the square cell around each fuel rod by a circular cell of equal area, calling it the equivalent cell (Fig. 3).

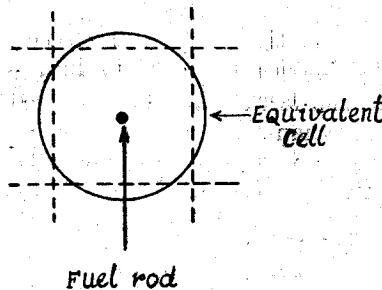


Fig. 3.

Let  $P$  be the probability that a primary fission neutron makes a collision in the fuel rod in which it is created and we denote by  $\sigma_f$ ,  $\sigma_c$ ,  $\sigma_i$  and  $\sigma_e$ , the fission, the non-fission capture, the inelastic scattering and the elastic scattering cross-sections respectively. the total cross section is

$$\sigma = \sigma_f + \sigma_c + \sigma_i + \sigma_e \quad \dots \quad (3)$$

Inside the rod, in the first collision,

- the number of fission neutrons produced .. .. .  $= \frac{\nu P \sigma_f}{\sigma}$
- the number of elastic collisions .. .. .  $= \frac{P \sigma_e}{\sigma}$ ,
- the number of neutrons escaping from the rod without collision  $= 1 - P$
- the number of neutrons slowed down below the  $U^{238}$  fission threshold in inelastic collision .. .. .  $= \frac{P \sigma_i}{\sigma}$ ,

where  $\nu$  is the average number of fission neutrons produced when  $U^{235}$  undergoes fission by capturing a thermal neutron.

If  $P'$  is the probability that second and higher generation fission neutrons will make a collision in the rod, then one can readily show that

$$\begin{aligned} \epsilon &= 1 - P + \frac{P \sigma_i}{\sigma} + P \frac{\nu \sigma_f + \sigma_e}{\sigma} \left( 1 - P' + \frac{P' \sigma_i}{\sigma} \right) + \\ &= 1 + \frac{\left[ (\nu - 1) - \frac{\sigma_e}{\sigma_f} \right] \frac{\sigma_f}{\sigma} P}{1 - P' \left( \frac{\nu \sigma_f + \sigma_e}{\sigma} \right)} \quad \dots \quad (4) \end{aligned}$$

$P$  and  $P'$  depend upon the dimensions of the rods and can be evaluated. For a rod of radius,  $a = 1.15$  cm,  $\epsilon = 1.025$ .

(iii) *Resonance Escape Probability.* Let us now consider how the resonance escape probability,  $p$ , is determined. To understand the nature of this factor we have to introduce a few new variables. By  $q(E)$ , called the slowing down density, we denote the number of neutrons slowing down past energy  $E$  per sec., per unit volume, and by  $\xi$  we denote the average of the logarithm of the ratio of final to initial neutron energy in any collision with a moderator nucleus. Further, we use the continuous slowing down theory of Fermi, which is a good approximation for heavy moderators like graphite and beryllium. According to this theory a neutron is assumed to lose energy at a continuous rate. The decrease in the logarithm of neutron energy per unit time is given by the product of  $\xi$  the average logarithmic neutron energy loss in a moderating collision, and the number of collisions a neutron makes in unit time,

$$-\frac{d}{dt} \log E = \xi \frac{v}{\lambda_s} \quad \dots \quad (5)$$

where  $\lambda_s = \frac{1}{N_1 \sigma_s}$ , is the scattering mean free path of neutron and  $v$  is its velocity.  $N_1$  is the number of moderating nuclei per unit volume.

Now the average number of neutrons  $n(E)dE$ , in any energy interval  $dE$  about  $E$ , would be equal to the number of neutrons that cross the energy

interval  $dE$  per sec. multiplied by the average time for which a neutron lingers in the interval. Thus

$$n(E)dE = q(E)dt$$

$$\text{or from (5)} \quad \phi(E) = n(E)v = \frac{q(E)}{\xi E} \quad \dots \quad (6)$$

$\phi(E)$  being the flux of neutrons of energy  $E$ .

If we assume that the resonance absorption in uranium at energy  $E$  is small, so that it does not produce any appreciable change in the flux at energy  $E$ , then we have

$$-dq = \sigma_{ao} N_o \phi(E) V_o/V_1 dE, \quad \dots \quad (7)$$

where  $N_o$  is the number of fuel atoms per unit volume,  $\sigma_{ao}$  the absorption cross section of uranium and  $V_o$  and  $V_1$  are the volumes in the cell taken up by uranium and graphite respectively. From (6) and (7)

$$-dq = \frac{\sigma_{ao} N_o V_o q}{\sigma_{s1} N_1 V_1 \xi E} dE$$

$$\text{or } p(E) = \frac{q(E)}{Q} = \exp \left\{ - \int_E^{E_o} \frac{V_o N_o \sigma_{ao}}{V_1 N_1 \sigma_{s1} \xi} \frac{dE'}{E'} \right\} \quad \dots \quad (8)$$

where  $Q$  is the source strength. The above result is based upon the assumption that the flux inside the fuel rod is constant, which is of course not correct. A correction for the depression of flux inside the fuel rod has to be made semi-empirically. For most cases of interest the numerical value of  $p$  is of the order of 0.9.

(iv) *Thermal Utilization Factor.* In order to get  $k_\infty$  we have still to determine the thermal utilization factor  $f$ . From the definition it follows that

$$\frac{1}{f} - 1 = \frac{\text{number of thermal neutrons captured in materials other than uranium}}{\text{number of thermal neutrons captured in uranium.}}$$

$$= \frac{V_1 N_1 \sigma_a \bar{\phi}_1}{V_o N_o \sigma_{ao} \bar{\phi}_o} \quad \dots \quad (9)$$

$\bar{\phi}_o$  and  $\bar{\phi}_1$  are the average thermal flux in uranium and graphite respectively. A rough estimate of  $\bar{\phi}_1/\bar{\phi}_o$  can be made by calculating the thermal neutron flux distribution in the cell using the classical diffusion theory. In actual cases  $f$  is nearly equal to 0.9.

From the four factor formula (1) we can now get the value of  $k_\infty$ . The maximum value of  $k_\infty$  that can be obtained for a graphite moderated natural uranium reactor is about 1.08. In order to obtain a self sustained chain reaction, the size of the reactor should be such, that for it, the non-leakage probability  $P$  is at least  $1/k_\infty$ .

The non-leakage probability  $P$  is a product of two factors, one being the probability of non-leakage of neutrons during slowing down and another the probability of non-leakage of thermal neutrons while diffusing. It will be convenient and simpler to consider a homogeneous assembly for evaluating  $P$ . We shall take account of the heterogeneity of the lattice in the final result.

(v) Let us first consider the non-leakage probability during the slowing down of neutrons in the reactor. Assuming the continuous slowing down theory of Fermi, we introduce (5) in the time dependent diffusion equation and solve it for  $q(E)$ , the slowing down density. We find that the number of neutrons thermalized per sec. per unit volume at any point  $r$  is given by

$$q(r_m, E_{th}) = k \infty \sigma_a N_0 \phi(r) \varepsilon^{-B^2 L_s^2} \dots \dots (10)$$

where  $L_s$  is the slowing down length and  $B$ , a constant depending upon the properties of the multiplying medium  $\phi(r)$  is the thermal flux at point  $r$ . For an infinite lattice there is no leakage of neutrons and hence

$$q_{\infty}(r, E_{th}) = \phi(r) N_0 \sigma_a \eta \varepsilon f p \dots \dots (11)$$

Therefore the non-leakage probability is  $\varepsilon^{-B^2 L_s^2}$ . For small values of  $B L_s$ , this reduces to  $1/(1+B^2 L_s^2)$ .

In the heterogeneous case, the slowing down length is increased on account of the fact that no slowing occurs in the uranium rods. The slowing down length for this case is obtained from the semi-empirical formula

$$L_s^2 = \frac{V_{total}}{V_1} (387 - 90Z) \text{ cm}^2$$

where  $Z$  is some given function of the fuel rod radius. In pure graphite  $L_s^2 = 387 \text{ cm}^2$  whereas in a cell of radius  $b = 9.0 \text{ cm}$  and uranium rod radius  $a = 1.15 \text{ cm}$ ; it is  $L_s^2 = 392 \text{ cm}^2$ .

(vi) In the homogeneous case, thermal neutron flux satisfies the equation

$$\nabla^2 \phi + B^2 \phi = 0 \dots \dots (12)$$

In order that (12) may have a solution in cylindrical geometry, we must have

$$B^2 = \left( \frac{2.405}{R} \right)^2 + \left( \frac{\pi}{H} \right)^2, \dots \dots (13)$$

which represents the lowest eigen value of  $B^2$ . Here  $R$  is the radius of the cylinder and  $H$  is its height. The number of neutrons moving out of a unit volume per sec is

$$-D \nabla^2 \phi = DB^2 \phi$$

$D$  being the diffusion coefficient. On the other hand, the number of thermal neutron absorbed per sec. per unit volume is  $N \sigma_{\infty} \phi(r)$ , so that

$$\frac{\text{Neutrons absorbed}}{\text{Neutrons absorbed} + \text{thermal leakage}} = \frac{1}{1 + B^2 L^2}$$

where  $L = \left( \frac{D}{N \sigma_a} \right)^{\frac{1}{2}}$ , is the "diffusion length" of thermal neutrons.

Thus  $1/(1+B^2 L^2)$  is the probability that a thermal neutron will not leak out of the system. In the heterogeneous case,

$$L^2 = 2500 \frac{V_t}{V_1} C_1 \text{ cm}^2$$

where  $C_1$  is the fraction of thermal neutrons captured by graphite.  $L^2$  in pure graphite is  $2500 \text{ cm}^2$ , whereas in a heterogeneous assembly it decreases to  $L^2 = 254 \text{ cm}^2$  ( $a = 1.15 \text{ cm}$ ;  $b = 9.0 \text{ cm}$ )

The total non-leakage probability is

$$P = \frac{1}{1+B^2 L_s^2} \cdot \frac{1}{1+B^2 L^2} = \frac{1}{1+B^2 M^2} \dots \dots (14)$$

where  $M^2=L^2+L_s^2$ . Combining (14) and (2), the condition for criticality of a finite reactor becomes 
$$\frac{k_\infty - 1}{M^2} = B^2 \quad \dots \quad (15)$$

If we consider a reactor in the form of a cylinder with height equal to the diameter, then using (13) we can rewrite (15) as

$$\frac{k_\infty - 1}{M^2} = \left(\frac{2.405}{R}\right)^2 + \left(\frac{\pi}{2R}\right)^2 \text{ or } R = 2.87 \left(\frac{M^2}{k_\infty - 1}\right)^{\frac{1}{2}} \quad \dots \quad (16)$$

In case the reactor is in the form of a cube, as is usually the case in practice—we assume that equation (15) still holds. However, instead of  $B^2$  being defined by (13) it is now given by

$$B^2 = 3 \left(\frac{\pi}{A}\right)^2 \quad \dots \quad (17)$$

where  $A$  is the side of the cube. Combining (17) and (15) we have

$$A = \pi \sqrt{3} \left(\frac{M^2}{k_\infty - 1}\right)^{\frac{1}{2}} \quad \dots \quad (18)$$

Since the various quantities on the r.h.s. of equation (16) or (18) are already known we can determine the radius  $R$  or side  $A$  that will make the reactor just critical. The critical side (radius) of the reactor comes out to be of the order of 5.7 meters (3 meters), for optimum values of  $a$  and  $b$ . However a reactor can be made critical with a smaller core provided the leakage of neutrons can be reduced. This is achieved in practice by surrounding the core with pure graphite which acts as a reflector, reflecting back almost 90 per cent of the neutrons that enter it. A detailed study shows that the core side or diameter can safely be reduced by an amount equal to twice the diffusion length  $L$  of thermal neutrons in the reflector material by using a reflector thickness of about  $\frac{3}{2}L$ . With a graphite reactor all round, of about 75 cm. thickness, the critical core side or diameter is reduced by almost one meter.

**Neutron Economy**

The values of  $p$  and  $f$  are too small in the case of a homogeneous assembly of uranium and graphite to permit self sustained chain reaction in the assembly. Only by taking uranium in the form of rods and surrounding them by graphite can one obtain  $p$  and  $f$  sufficiently large to give  $k_\infty \gg 1$ . To illustrate that even in the heterogeneous case the margin within which the various factors can be varied is very small, we give in the following table what is called the neutron balance sheet.

**TABLE**

Neutrons from $U^{235}$ fission	..	..	..	..	..	2.5
Fast fission neutrons from $U^{238}$	..	..	..	..	..	0.06
Total fission neutrons						2.56
Neutrons to maintain chain reaction	..	..	..	..	..	1.00
Neutrons absorbed in $U^{238}$ to make $Pu^{239}$	..	..	..	..	..	0.90
Neutrons absorbed in $U^{235}$ to make $U^{236}$	..	..	..	..	..	0.20
Neutrons absorbed in Moderator	..	..	..	..	..	0.30
Neutrons absorbed in structural materials	..	..	..	..	..	0.05
Neutrons escaping from core	..	..	..	..	..	0.09
Total neutron expenditure						2.54
Excess neutrons						0.02

It is seen that excess neutrons are even less than 1 per cent of the fission neutrons, and hence strict neutron economy has to be observed in building a reactor. The economy can be achieved on several scores. Firstly by using as structural materials, metals like aluminium, magnesium, zirconium, which have a low absorption cross-section for thermal neutrons. Then the graphite for the moderator should be of nuclear purity. Boron, which is the main impurity in graphite cannot be tolerated above 0.5 part in a million. The coolant, which is used to cool the uranium rods in the reactor should not absorb neutrons heavily. Finally, the reactor should be built as an optimum lattice.

**Reactor Control**

A reactor is always built larger than the critical size. The rate at which the neutron density builds up in such a reactor is given by

$$n = n_0 e^{(k-1)t/l} \dots \dots \dots (19)$$

where  $l$  is the average life time of neutrons in the reactor, and  $n_0$  is the initial number of neutrons. In case we neglect the effect of a few delayed neutrons that always accompany fission of  $U^{235}$ , the life time of a neutron comes out to be of the order of 0.001 sec. It follows from (19) that even for  $k-1 = 0.01$ , the number of neutrons would increase by factor of  $2 \times 10^4$  every sec. Such a reactor would be impossible to control. Luckily the effect of delayed neutrons, which are only about 0.75 per cent of the total number of fission neutrons, is to increase the average life time of neutrons to  $l = 0.1$  sec. For the same excess reactivity  $k-1 = 0.01$ , the neutrons would now increase only by a factor of 2.7 every 10 secs. It is thus the presence of these delayed neutrons in fission that makes a controlled chain reaction possible.

In a reactor, the chain reaction is controlled by moving cadmium or boron steel rods in or out of it. On account of very high thermal neutron absorption cross-section of cadmium and boron, they can effectively control the reactivity and hence the thermal flux. The chain reaction can be stopped by pushing in the rods sufficiently, so that more neutrons are absorbed per sec. than are produced. Under such conditions the flux soon goes down almost to zero.

**Power**

The power level of a reactor is related in a simple way to the maximum thermal flux, by the formula

$$\text{power (K.W.)} = \frac{\sigma_f N_o \phi_{\text{max}} V_o \times 200 \times 10^6 \times 1.6 \times 10^{-12}}{10^{10}} \times \frac{1}{2} \times \frac{1}{1.6} \dots (20)$$

where we have assumed that the flux in uranium rod is about 1/2 that in graphite and that the average flux is about 1/1.6 of the maximum flux. From the above relation it follows that

$$\phi_{\text{max}} = 1.1 \times 10^{10} \frac{\text{Power extracted in K.W.}}{\text{ton of Uranium}} \dots \dots \dots (21)$$

Using compressed gas to cool uranium rods one can extract up to 1000 kw per ton of uranium, giving a maximum thermal flux in a graphite moderated reactor as  $\sim 10^{13} \text{ cm}^{-2} \text{ sec}^{-1}$ .



The power level at which a reactor works determines the life of fuel rods. For 1 Megawatt day of energy extracted, 1 gm of  $U^{235}$  is used up. Depletion of  $U^{235}$  and the increase of fission products like xenon 135 and samarium 149, which have a large thermal neutron absorption cross-section decreases the reactivity  $\frac{k-1}{k}$  of the reactor. Depending upon the excess reactivity that is built in at the start, the above factors determine the time when the fuel rods are to be replaced.

### Description of a Reactor

To get a better idea of the magnitudes of various quantities involved we shall briefly describe an actual reactor. As an example we take the Oak Ridge National Laboratory Graphite Reactor (ORNL Graphite Reactor or X-pile) (Fig. 4) which first became critical on 3rd November 1943. The

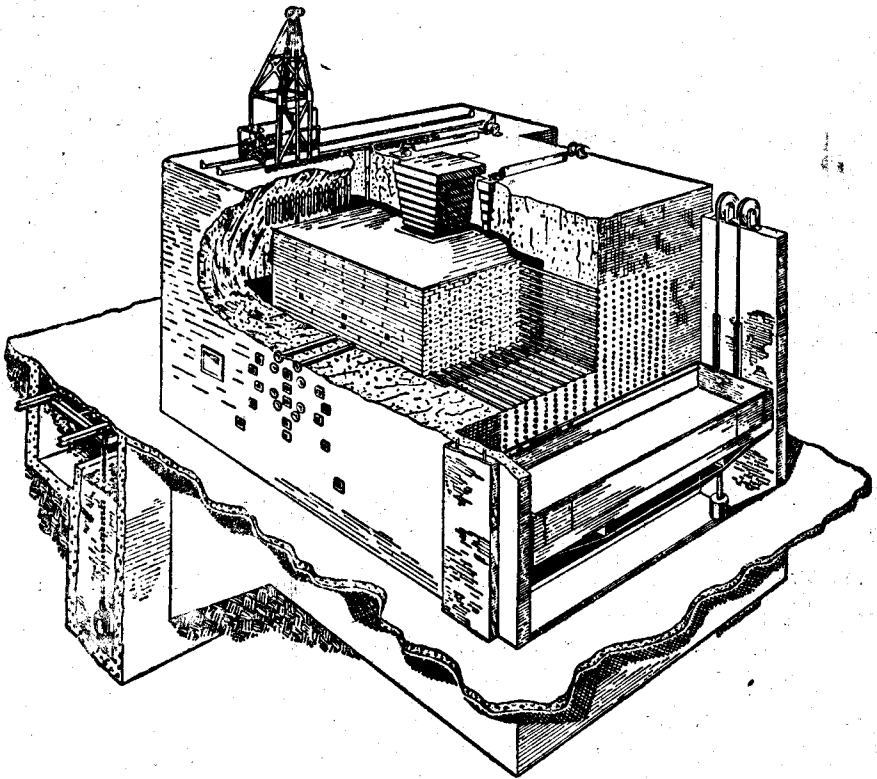


Fig. 4. Oak Ridge X-Pile

external dimensions of the reactor are  $38' \times 47' \times 35'$ . The moderator is a  $24'$  cube of graphite built out of blocks of size  $4'' \times 4'' \times 50''$ . Some blocks have V-cuts in them, so that when assembled, form diamond shaped holes running horizontally all the way through the moderator. There are 1248

holes spaced with centres 8" apart. These are the fuel channels of the reactor. There are other holes both horizontal and vertical, left in the reactor to take out neutrons beams for experimentation.

A "biological shield" of 7' thick concrete is there to prevent neutrons and  $\gamma$ -radiation from escaping out. Between the shield and the moderator on the two sides of the fuel channels, are air gaps which serve as inlet and outlet for cooling air. Since the temperature of the outgoing air is not very high, it is of no use as far as power generation is concerned, and is discharged into the atmosphere through a high chimney after being filtered for radio-active dust.

The uranium "slugs" are slightly over 1" in diameter and 4" long, each piece weighing about  $2\frac{1}{2}$  lbs. The rod has an aluminium canning or "sheath" of 35/1000" thickness. This is essential to prevent highly radioactive fission products from escaping and also to prevent the oxidation and corrosion of uranium.

There are five safety rods in the reactor. Three are cadmium rods encased in steel and are 8' long. These are supposed to fall vertically under gravity in an emergency. Two other rods of boron steel can be pushed in horizontally under hydraulic pressure in case of trouble. The reactor is controlled by two boron steel rods that are driven electrically.

Total mass of graphite used is about 600 tons (price of machined graphite for moderator  $\sim$  Rs. 10,000 per ton) and that of uranium is about 35 tons (price  $\sim$  Rs. 350,000 per ton).

Maximum thermal neutron flux is nearly  $1.2 \times 10^{12}$  and the reactor works at a power level of  $\sim$  3,800 k.w.

### Problems connected with a reactor

Mention of a few of the problems that face a reactor engineer may not be out of place here. Canning of uranium rods presents one of the difficult problems. The sheath which is usually of aluminium has to be sufficiently thin so that the neutron economy is not too adversely affected. It should also fit the uranium rod tightly so that heat from the rod can be efficiently removed. Failure of a sheath even to the extent of a tiny hole requires the reactor to be shut down and the rod replaced immediately for fear of highly radioactive fission products being carried out of the reactor.

Inside the reactor uranium, moderator and other structural materials are under intense neutron bombardment for long periods of time. These produce dislocations in the materials and consequently their physical properties are changed. The nature of the changes is not at all well understood.

Cooling the fuel rods specially in power reactors is another of the important problems. The coolant should have very low neutron absorption cross-section and at the same time it must have very good thermal properties. The cooling problem is aggravated by the fact that uranium has very low thermal conductivity and besides the temperature at any point of the

rod cannot be allowed to go above  $600^{\circ}\text{C}$ , as little above this temperature there is a phase transformation in uranium.

Safe disposal of the highly radioactive fission products presents another problem.

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