

## Nuclear Theory - Course 127

## MODERATOR PROPERTIES

In the previous lesson you learnt that it is possible to sustain a chain reaction either by enriching the fuel in U-235, or by adding a moderator to natural uranium fuel.

Up till now, the second alternative has been by far the most popular one, and the major differences in the design of power reactors have, in the first instance, been dependent on the type of moderator chosen. In this lesson we shall examine the basis on which this choice is made. In other words, this boils down to looking at the requirements of a moderator and then seeing to what extent these are met by the various possible moderator materials.

Neutron Absorption

The primary objective of a moderator is a lot easier to express than to achieve: the fission neutrons must be slowed down to thermal energies without being absorbed. Let us examine the latter aspect first:

There are two possibilities:- the neutrons can be absorbed by the moderator atoms themselves or by fuel atoms, and this can occur anywhere in the energy range from  $\sim 2\text{MeV}$  (fission neutrons) down to  $0.025\text{ eV}$  (thermal neutrons). Absorption by moderator atoms can obviously be minimized by choosing a moderator with a sufficiently low absorption cross section, but for fuel the argument is rather more subtle.

If you refer back to Fig. 2 of the previous lesson, you will see that U-238 exhibits a number of severe  $(n,\gamma)$  absorption peaks between 5 and 100 eV. These are called *resonances*, and any neutrons absorbed by them are said to have suffered *resonance capture*. It is essential to minimize such resonance capture, and one way of doing this is to ensure that, in the slowing down process, the neutron energy loss per collision is as high as possible. For example, consider the two figures on the following page (for the sake of simplicity the resonances have been smoothed out).

Moderator 2 thermalizes the neutrons in far fewer collisions than moderator 1. This means that the neutrons in moderator 2 will spend less time in the resonance energy region, and will therefore also have less chance of colliding with U-238 while they have this energy. If they do, they will almost certainly be captured, and if you don't believe this have another look at Assignment #2 in the earlier lesson in cross sections. The conclusion of all this then is that there will be less resonance capture in U-238 with moderator 2 than with moderator 1.

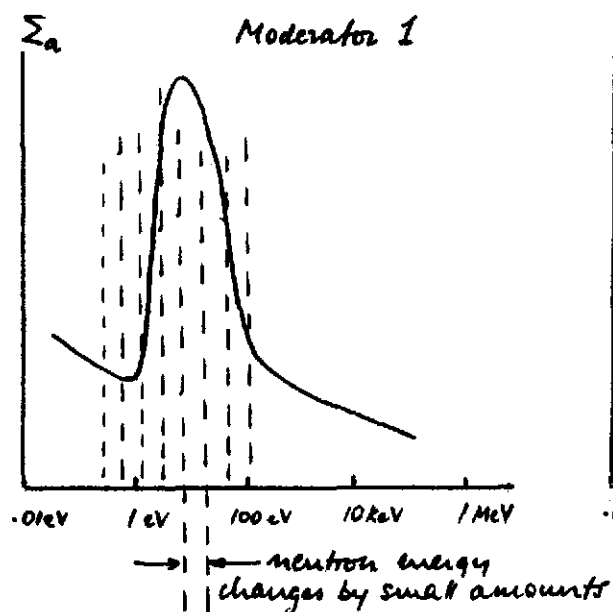


Fig. 1.

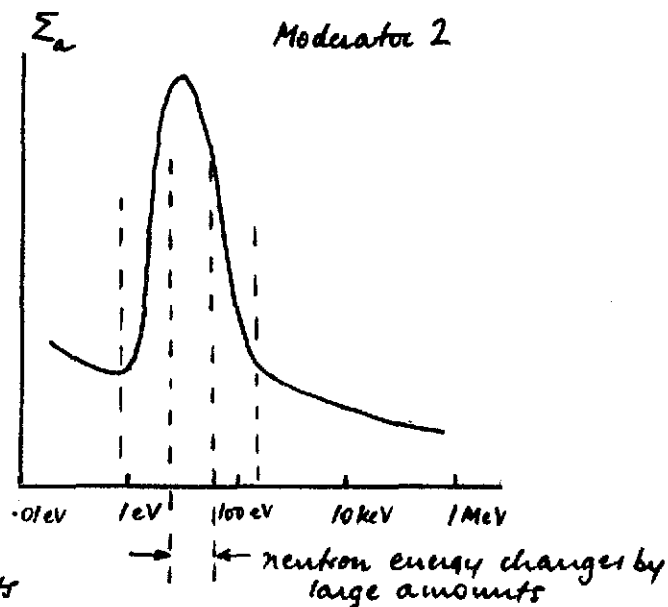


Fig. 2.

### Effect of Moderator on Resonance Capture

Apart from this, during the slowing down process there will be less collisions and hence less neutron absorption by the moderator atoms themselves if we use moderator 2 rather than 1.

### Slowing Down Mechanism

Having established that we want to slow the neutrons down in as few collisions as possible we shall now examine how this might be achieved.

There are two slowing down mechanisms:

- (1) inelastic scattering (with fuel nuclei)
- (2) elastic scattering (with moderator nuclei)

(Inelastic scattering with moderator nuclei is not possible because the energies are too low, and even with uranium nuclei it is only possible down to about 100 keV. In any case, it is relatively unimportant, as we shall see later. Elastic scattering with fuel nuclei may be ignored, because there the energy loss per collision is negligible).

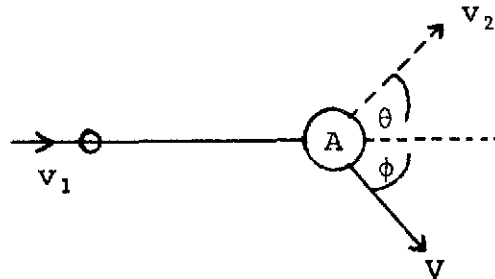


Fig. 3. Elastic Collision of Neutron with Moderator Nucleus

In an elastic collision the energy loss  $\Delta E$  of a neutron of initial energy  $E$  is given by:

$$\frac{\Delta E}{E} = \frac{4A \cos^2 \phi}{(A+1)^2} \quad \text{—————} \quad (1)$$

This expression shows that the fractional energy loss depends both on the incident angle  $\phi$  and on the mass of the moderator nucleus. For many collisions the energy loss must be weighted according to the probability of a collision occurring at an angle  $\phi$ , and a weighted mean energy loss per collision can then be found.

Now consider that  $N$  collisions with moderator nuclei are required to slow the fast neutrons (energy  $E_1$ ) down to thermal values (energy  $E_{th}$ ). The energy after the first impact is  $E_2$ , after the second  $E_3$ , and so on. We can therefore say that:

$$\frac{E_1}{E_{th}} = \left( \frac{E_1}{E_2} \right) \times \left( \frac{E_2}{E_3} \right) \dots \dots \left( \frac{E_i}{E_{i+1}} \right) \dots \dots \left( \frac{E_N}{E_{N+1}} \right) = \left( \overline{\frac{E_i}{E_{i+1}}} \right)^N$$

where  $\left( \overline{E_i/E_{i+1}} \right)$  signifies the average energy loss per collision.

Taking logs to base e gives

$$\ln \frac{E_1}{E_{th}} = N \log \frac{E_i}{E_{i+1}} = N\xi \quad (2)$$

$\xi$  is called the *mean logarithmic energy decrement*, and is given by

$$\xi = 1 + \frac{(A-1)^2}{2A} \ln \left( \frac{A-1}{A+1} \right) \quad (3)$$

(This expression can be obtained after some manipulation when the weighted mean of  $\cos^2\phi$  is applied to equation (1). It is not my intent to bore you with the details since it is only calculus).

Substituting  $E_1=2\text{MeV}$  and  $E_{th}=0.025\text{eV}$  into (2) gives us

$$\begin{aligned} \ln \frac{2 \times 10^6}{0.025} &= 18.2 = N\xi \\ \text{i.e. } N &= \frac{18.2}{\xi} \end{aligned} \quad (4)$$

This shows that a small number of slowing down collisions can only be achieved with a large value of  $\xi$ , or a small value of  $A$  since a fairly close approximation for  $\xi$  is

$$\xi \approx \frac{2}{A + \frac{2}{3}} \quad (5)$$

For  $A > 10$ , this approximation is good to within 1%.

At this point it might be constructive to consider the effect of inelastic scatter in the fuel. If we assume that this reduces the average neutron energy from 2MeV to perhaps 1 MeV, then equation (4) changes to

$$N\xi = \ln \frac{1 \times 10^6}{0.025} = 17.5$$

This means that the number of collisions the neutrons have to make to reach thermal energies is reduced to  $17.5/\xi$ , ie, by less than 4%. For this reason, any slowing down effects by inelastic scatter in the fuel are usually ignored, and the number of collisions to thermalize is still taken as  $18.2/\xi$ .

If the moderator is not a single element, but a compound such as  $D_2O$ , the effective value of  $\xi$  is given by

$$\xi_{(D_2O)} = \frac{2\sigma_s(D)\xi(D) + \sigma_s(O)\xi_O}{2\sigma_s(D) + \sigma_s(O)} \quad (6)$$

Table 1 shows the accurate values of  $\xi$  (equation 3) of a number of light materials which might be suitable as moderators.

TABLE 1  
Mean Logarithmic Decrements

	$\xi$	Collisions to Thermalize
$H^1$	1.000	18
$H^2$	0.725	25
$He^4$	0.425	43
$Be^9$	0.206	83
$C^{12}$	0.158	115
$H_2O$	0.927	20
$D_2O$	0.510	36
$BeO$	0.174	105

#### Slowing Down Power and Moderating Ratios

A small number of collisions to thermalize is obviously desirable, but this is of no use on its own unless the collisions actually occur. This implies that  $\lambda_s$ , the mean free path for scattering collisions, must be small. Therefore

$$\Sigma_s = \frac{1}{\lambda_s} = N'\sigma_s$$

must be large. This immediately rules out gases as moderators, because  $N'$  would be too small for the neutrons to be slowed down within a reasonable distance.

The overall effectiveness of a material for slowing down neutrons is measured by the product  $\xi\Sigma_s$ , which is known as the *Slowing Down Power*. You should be able to show that it is the average decrease in the log of neutron energy per cm of path.

$\lambda$  = mean free path.

Table 2 shows the slowing down powers of the solid and liquid moderators introduced in Table 1. The value for helium is also shown to demonstrate the unsuitability of a gas.

TABLE 2  
Slowing Down Powers and Moderating Ratios

	$\xi$	$\Sigma_s \text{ (cm}^{-1}\text{) (a)}$	$\xi \Sigma_s$	$\Sigma_a$	$\xi \Sigma_s / \Sigma_a$
He (b)	0.425	$21 \times 10^{-6}$	$9 \times 10^{-6}$	? very small	? large
Be	0.206	0.74	0.15	$1.17 \times 10^{-3}$	130
C (c)	0.158	0.38	0.06	$0.38 \times 10^{-3}$	160
BeO	0.174	0.69	0.12	$0.68 \times 10^{-3}$	180
H <sub>2</sub> O	0.927	1.47	1.36	$22 \times 10^{-3}$	60
(100%) D <sub>2</sub> O	0.510	0.35	0.18	$0.33 \times 10^{-6} \text{ (d)}$	5500 (d)
O <sub>2</sub> O (reactor grade 99.75 %)					

- (a)  $\Sigma_s$  values of epithermal neutrons (ie, between  $\sim 1$  and  $\sim 1000$  eV)  
 (b) at S.T.P.  
 (c) reactor-grade graphite.  
 (d) 100% pure D<sub>2</sub>O.

Not only must the moderator be effective in slowing down neutrons, but it must also have a small capture cross section. Neutrons are slowed down to decrease radiative captures compared to fission captures, and obviously the whole purpose of moderation would be defeated if the moderator nuclei themselves captured neutrons.

A reasonable indication of the overall quality of a moderator is the *Moderating Ratio*, which combines the slowing down power and the macroscopic capture cross section:-

$$\text{Moderating Ratio} = \frac{\xi \Sigma_s}{\Sigma_a} \quad (7)$$

We are now in a position to draw some interesting conclusions from Table 2.

$H_2O$  has excellent slowing down properties, and is often used as a fast neutron shield (neutrons must be slowed down before they can be absorbed. Why?) Unfortunately its  $\Sigma_a$  is too high to permit its use as a moderator for natural uranium fuel, and enrichment will be necessary.

Be, BeO and graphite have lower values of  $\Sigma_a$ , and can be used with natural uranium fuel provided it is in metal form. The use of natural uranium compounds with more attractive physical and chemical properties (such as  $UO_2$  or UC) is not feasible with these moderators, because of the reduction in the concentration of uranium atoms. The British line of power reactors used a graphite moderator with natural uranium metal fuel (their earlier Magnox stations), and in the early '60s they changed to graphite with enriched  $UO_2$  fuel (the AGR stations).

*Advanced Graphite Reactor.*

In the U.S., an abundance of U-235 and a tradition of using it in nuclear submarines led to all out development of light water reactors with relatively highly enriched fuel and a relatively poor moderator.

You can see from Table 2 that heavy water is by far the best moderator as far as its nuclear properties go, and of course its use was adopted for the CANDU line of reactors (CANDU = CANadian-Deuterium-Uranium). Its  $\Sigma_a$  is so low that natural uranium can even be used in compound form as  $UO_2$ .

The substance used as a moderator must be very pure. It is usually used, in a reactor, in larger amounts than any other material, eg, the volume of carbon in a graphite moderated reactor is 70 to 80 times that of the fuel. A very small amount of impurity in a moderator can substantially increase its capture cross section. The addition of 1 boron atom to every million graphite atoms would increase the capture cross-section of graphite by 25%.

For the same reason the isotopic purity of  $D_2O$  must be kept high. The addition of 0.25%  $H_2O$  to pure  $D_2O$  more than doubles the capture cross section. The isotopic purity of moderator  $D_2O$  is kept at 99.75% by weight or better. This is known as *reactor-grade  $D_2O$* . This isotopic purity is not easy to maintain since heavy water is hygroscopic, nevertheless it is imperative that all  $H_2O$  be excluded. For example, a 0.25% decrease in isotopic purity in Pickering would cause a reduction in reactivity equivalent to a fuel burn-up loss of 21 full power days. This represents an amount of money which is so high as to be almost outside the audible range of the human ear.

### The Diffusion of Neutrons Through the Moderator

Many parameters which determine the design of a reactor are dependent on the way neutrons are slowed down and diffuse in the moderator.

Neutrons diffuse through a material as a result of being scattered by nuclei. Neutrons virtually never collide with each other because the neutron density is so much smaller than the atomic density. The treatment of neutron diffusion, which is a process similar to the diffusion of electron in a metal, is too complicated to include in this course, and we shall therefore restrict ourselves to a pictorial representation.

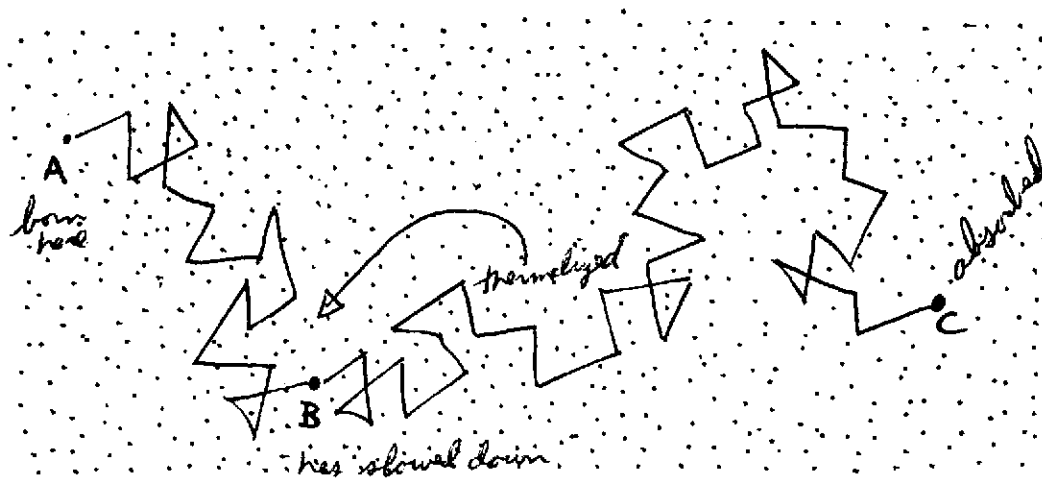


Fig. 4. Neutron Diffusion In A Moderator

A fission neutron born at A is thermalized in  $18.2/\xi$  collision to arrive at B. The mean square of the "crow-flight" distance AB (it always seems to be assumed that crows bomb along in straight lines) is given by the expression

$$6L_s^2 = \overline{(AB)^2} \quad (8)$$

$L_s$  is called the *slowing down length*, and it is an important parameter in reactor physics. For  $D_2O$   $L_s = 11$  cm, and hence  $\overline{(AB)^2}$  is about  $725 \text{ cm}^2$ , with  $AB$  around  $23 \text{ cm}^*$ . This can be compared with an average total distance travelled during slowing down, which is the number of collisions times the mean free path for elastic scatter. For  $D_2O$ , this turns out to be around  $100 \text{ cm}$ .

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\*) You realise that  $\overline{x^2} \neq \overline{x}^2$ , don't you?



After the neutron has been thermalized at B, it will zip around in the moderator before it is finally absorbed at C. A similar relationship again applies, namely

$$6L^2 = \overline{(BC)^2}, \quad (9)$$

where  $L$  is the *diffusion length*. For  $D_2O$  this is very large at 100 cm, and you will probably realise that this reflects the exceptionally low  $\Sigma_a$  of  $D_2O$ .

(In fact, other things being equal,  $L^2$  is inversely proportional to  $\Sigma_a$ .)

TABLE 3

Slowing Down and Diffusion Lengths

Moderator	$L_s$ (cm)	$L$ (cm)
$H_2O$	5.6	2.76
$D_2O^*$	<u>11.0</u>	<u>100</u>
Be	9.2	21
Graphite	18.7	64.2

\* reactor-grade

The slowing down length determines the optimum distance between adjacent fuel channels of a heterogenous reactor. This distance is called the *lattice pitch*, and the values of Table 3 should explain why  $H_2O$  moderated reactors are smaller than  $D_2O$  moderated reactors.

The low value of  $L$  for  $H_2O$  reflects its high  $\Sigma_a$ , and you can see that *overmoderating* a light water reactor (ie, having a greater lattice pitch than is needed) would carry such a severe penalty in terms of neutron absorption that it is never contemplated. On the other hand, overmoderating a heavy water reactor by a few cm makes very little difference to the absorption. In this case the designer might wish to increase the lattice pitch somewhat to give himself more flexibility for the design of end fittings and end shields. (All of the CANDU reactors are overmoderated - a pressure tube design makes it almost impossible to be otherwise.)

The values of  $L$  given in the table of course apply to a moderator alone, because it is assumed that the thermal neutrons are finally absorbed by moderator nuclei. In a reactor containing fuel, the large majority of thermal neutrons will be absorbed by fuel nuclei and the effective diffusion length is then considerably reduced. The slowing down length should not be affected much because the contribution of the fuel nuclei to the slowing down process is negligible. For the sake of comparison, the values for the Pickering cores are  $L_s^2 = 145 \text{ cm}^2$  and  $L^2 = 224 \text{ cm}^2$ .

A final point is that the fraction of neutrons escaping from the core, called the *leakage*, also depends on  $L^2$  and  $L_s^2$ . We shall leave the discussion of this to the ~~next~~ lesson.

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### ASSIGNMENT

1. Why is  $\xi_{\text{H}_2\text{O}}$  so much closer to  $\xi_{\text{H}}$  than  $\xi_{\text{D}_2\text{O}}$  is to  $\xi_{\text{D}}$ ?
2. How many collisions are required for neutrons to lose, on average, 99% of an initial energy of 2 MeV in  $\text{D}_2\text{O}$ ? Compare this with the total number of collisions required to reach thermal energies.
3. Calculate the total zig-zag path made by fission neutrons slowing down to thermal energy in Helium gas. Compare this with the corresponding distance in  $\text{H}_2\text{O}$ .
4. Calculate the moderating ratio for reactor-grade  $\text{D}_2\text{O}$ , and insert this value in Table 2.
5. The time taken by a neutron to slow down from energy  $E_0$  to Energy  $E_t$  is given by

$$t_s = \frac{\sqrt{2m}}{\xi \Sigma_s} \left( \frac{1}{\sqrt{E_t}} - \frac{1}{\sqrt{E_0}} \right) \quad (m = \text{neutron mass})$$

Calculate the slowing down times for  $\text{H}_2\text{O}$ ,  $\text{D}_2\text{O}$  and graphite. Can you derive this expression?

6. For neutrons that have just been thermalized in reactor-grade  $\text{D}_2\text{O}$ , calculate the total zig-zag path, the number of collisions and the survival time before capture.
7. Explain why the values of  $L_s$  and  $L$  for the Pickering cores differ from those of  $\text{D}_2\text{O}$ .

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