

Module 7

Effects of Fuel Burnup

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7.1 MODULE OVERVIEW

While a reactor is operating, neutron reactions are continuously changing the composition of the core. U-235 is being burnt up, Pu-239 and Pu-241 are being created and Pu-240 and fission product poisons are building up. This has obvious operational consequences, since the reactivity of the system must be continuously adjusted, for example by changing the boron content of the moderator, to keep the reactor critical.

In this module, we will discuss the principal changes that are occurring and examine their magnitude and the time scale on which they are taking place. We start by defining a *unit*, the *neutron per kilobarn*, which we will use to specify the degree of fuel burnup. We will then look specifically at the rates at which U-235 is consumed and Pu-239 builds up as a function of time at full power. These studies will enable us to consider how the *overall reactivity* of the reactor varies as irradiation proceeds, which in turn determines the reactivity required to maintain criticality. We will also briefly consider the effect of fuel burnup on reactor kinetics. The final section of the module will review some practical aspects of fuel management.

7.2 MODULE OBJECTIVES

After studying this module, you should be able to:

- i) State and explain the units used for fuel burnup.
- ii) Write the equation for the burnup rate of U-235, and its solution, and roughly sketch the curves for U-235 concentration as a function of time.

- iii) Write the equation illustrating the Pu-239 increase rate, and its solution, and roughly sketch the curve for Pu-239 buildup.
- iv) Explain why the Pu-239 concentration tends towards an equilibrium value as the fuel burnup increases.
- v) Draw a rough graph showing how overall reactor reactivity changes with time at full power, and explain the shape of the graph in terms of the main components which contribute to it.
- vi) Explain how and why the four factors in the formula for k_{∞} change with burnup.
- vii) Explain why reactor kinetics are affected by the degree of fuel burnup.
- viii) List some of the factors a fuel engineer must consider when working out a refuelling program.

7.3 FUEL BURNUP – GENERAL

In a freshly-fuelled core, the only fissile material is the U-235 which constitutes 0.72% of the natural uranium. As the reactor operates, the U-235 is gradually depleted, lessening reactivity; a further loss is incurred by the buildup of the fission products which have significant absorption cross-sections for thermal neutrons. This is, however, partially compensated by the buildup of fissile Pu-239 following neutron capture in the U-238. The gradual change in fuel composition produces three principal effects:

- i) long-term reactivity changes.

- ii) changes in the kinetic behavior of the reactor (see Module 8).
- iii) changes in the neutron flux distribution (already mentioned in our discussion of bi-directional refuelling and differential burnup).

Before reviewing the effects produced by fuel burnup, we should look at how the *degree of burnup* of fuel is specified. The two main units in common use are described below.

a) **Energy extracted per unit mass**

As stated in Section 2.8.7, since each gram of U-235 burnt up produces approximately one megawatt-day of energy in the form of heat, one way to specify burnup is in terms of the total cumulative heat energy extracted per unit mass of fuel. The unit we will use is *megawatt-hours per kilogram uranium* (MWh/kgU). Note that the MWh here are thermal, not electrical, energy.

b) **Total neutron exposure**

Since the rate of fissioning is given by the product $\phi\Sigma_f$ per unit volume (Section 3.6), the rate of burnup is proportional to the neutron flux. The accumulated burnup over a specific period of time (t) is therefore proportional to the product of flux and time (ϕt). This product is known as the *total neutron exposure* of the fuel. The units of neutron exposure are

$$\phi t \rightarrow \frac{\text{neutrons} \times s}{\text{cm}^2 s} \rightarrow \text{neutrons} / \text{cm}^2$$

A commonly used modification of the unit is the so-called *neutron per kilobarn*, defined by changing the unit of area to the kilobarn:

$$1 \text{ kb} = 10^3 \text{ b} = 10^3 \times 10^{-24} \text{ cm}^2 = 10^{-21} \text{ cm}^2$$

Burnup units

MWh/kgU

Total neutron exposure (ϕt)

Neutron per kilobarn (n/kb)

The relation between n/kb and n/cm^2 is then:

$$\phi t = 1 n / kb = \frac{1 \text{ neutron}}{10^{-21} \text{ cm}^2} = 10^{21} \text{ neutrons / cm}^2$$

One way of looking at this rather obscure unit of exposure is to say that 1 n/kb is equivalent to 10^{21} neutron-cm per cm^3 , that is, the accumulated track length of all neutrons that have passed through a unit volume of the fuel during its irradiation. The typical average exit burnup for a CANDU reactor is around 1.8 n/kb .

The approximate relationship between the two units of burnup measurement for the freshly-fuelled core is:

$$1 \text{ n/kb} \approx 100 \text{ MWh/kgU}$$

Another way one can specify burnup is by the number of equivalent full-power days (EFPD), that is, the number of days the reactor would have to operate at full power to achieve a given burnup in MWh/kgU. The relation between EFPD and the burnup units already mentioned for a CANDU with a full-power average flux of $10^{14} \text{ n/cm}^2\text{s}$ may be stated with a reasonable degree of accuracy as

$$10^{21} \text{ neutrons/cm}^2 = 1 \text{ n/kb} = 100 \text{ MWh/kgU} = 115 \text{ EFPD}$$

Equivalent full power days (EFPD)

7.4 LONG-TERM REACTIVITY EFFECTS

We will review the rates of burnup of U-235 and buildup of Pu-239, and the overall rate of reactivity change in the reactor due to the changing composition of the fuel.

7.4.1 Burnup Rate of U-235

The rate at which U-235 is destroyed by neutron absorption per unit volume is given by the product $\Sigma_{a5}\phi$. This is expressed as

$$\frac{dN_5}{dt} = -\Sigma_{a5}\phi = -N_5\sigma_{a5}\phi \quad (7.1)$$

where

N_5 is the number of U-235 nuclei *per cm³* at a specific time,

σ_{a5} is the microscopic *absorption* cross-section of U-235 (in cm^2),

ϕ is the neutron flux in $\text{n/cm}^2 \text{ s}$.

The negative sign on the right hand side takes account of the fact that N_5 is decreasing.

This equation is obviously of the same form as the radioactive decay equation:

$$\frac{dN}{dt} = -\lambda N$$

where the quantity equivalent to the decay constant λ is the product ($\sigma_{a5}\phi$) in the present case.

Burnup rate of U-235

The *solution* to equation 7.1 is therefore of the same form as in the radioactivity case, i.e.,

$$N_5(t) = N_{50}e^{-\sigma_{a,5}\phi t} \quad (7.2)$$

where N_{50} is the number of U-235 nuclei/cm³ at $t = 0$.

The U-235 concentration will therefore decrease in exponential fashion, as illustrated in Figure 7.1.

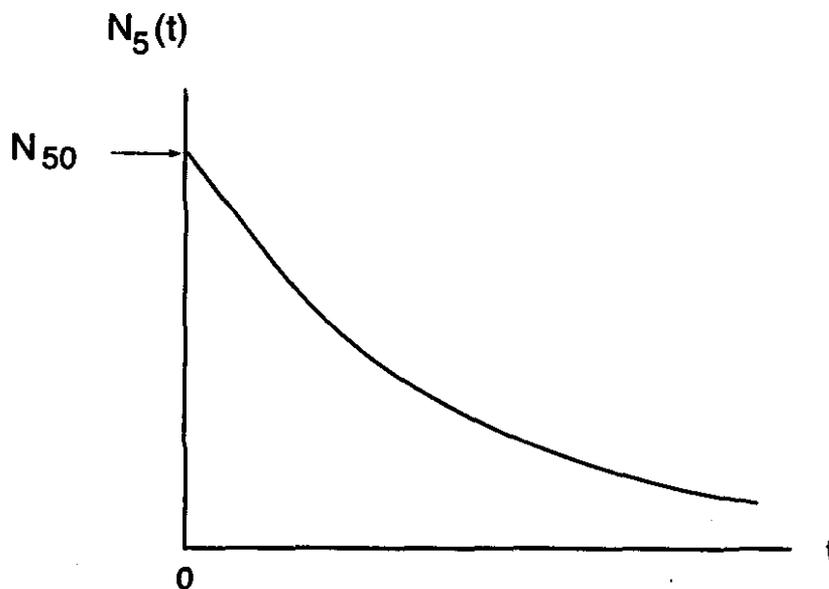


Figure 7.1: Exponential decrease in U-235 concentration

7.4.2 Buildup Rate of Pu-239

Pu-239 is formed as a result of neutron capture by a U-238 nucleus, followed by two successive beta decays (see Section 2.7). The rate at which Pu-239 is formed per unit volume equals the capture rate in U-238, or

$$N_8 \sigma_{a8} \phi$$

where N_8 is the number of U-238 atoms/cm³ (which remains effectively constant since its rate of burnup is so small relative to the concentration; σ_{a8} , the absorption cross-section of U-238, is only 2.7 b).

The Pu-239 formed is destroyed in two ways:

1. by neutron absorption (causing fission or production of Pu-240)
2. by decay (Pu-239 is an alpha emitter).

In the sort of flux characteristic of a power reactor, the neutron absorption rate in Pu-239 is much greater than its radioactive decay rate. We can therefore ignore radioactive decay.

The rate of loss of Pu-239 (atoms/cm³s) then equals

$$N_9 \sigma_{a9} \phi$$

The net change in Pu-239 concentration per cm³ per sec is:

$$\frac{dN_9}{dt} = N_8 \sigma_{a8} \phi - N_9 \sigma_{a9} \phi \quad (7.3)$$

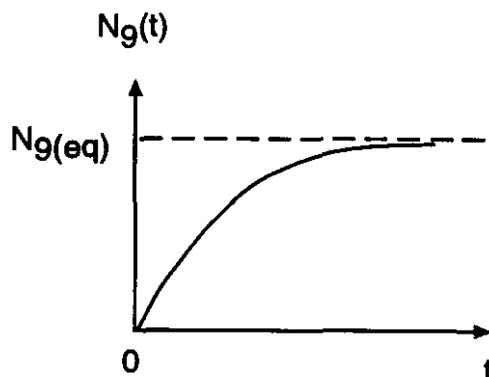


Figure 7.2: Build-up of Pu-239 toward equilibrium

The solution to this equation is

$$N_g(t) = \frac{N_g \sigma_{238}}{\sigma_{239}} [1 - e^{-\sigma_{239} t}] \quad (7.4)$$

The way the Pu-239 concentration builds up, according to this equation, is shown in Figure 7.2. Initially, the rate of build-up is relatively fast, because the second term in equation 7.3 is small. As N_g increases, however, the second term builds up, and the rate of increase of Pu-239 begins to slow down. Eventually, the second term becomes equal to the (constant) first term, and dN_g/dt reduces to zero. Once this has occurred, N_g remains constant, at a value known as the *equilibrium value*, $N_{g(eq)}$. We can calculate this equilibrium value either by taking $dN_g/dt = 0$ in equation (7.3) or by taking $t = \infty$ in equation (7.4). The equilibrium value is

$$N_{g(eq)} = \frac{N_g \sigma_{238}}{\sigma_{239}} \quad (7.5)$$

and equation 7.4 can then be written

$$N_p = N_{p(eq)} [1 - e^{-\sigma_{ap}\phi t}] \quad (7.6)$$

with $N_{p(eq)}$ given by equation 7.5.

Note that substituting $\sigma_{ab} = 2.7$ b and $\sigma_{ap} = 1013$ b in equation 7.5 gives $N_{p(eq)}/N_b = 0.27\%$. This may be compared to the initial U-235 fraction of 0.72%. In practice, due to the overall loss of reactivity from burnup, it is necessary to start replacing the fuel in the core well before the Pu-239 has reached equilibrium.

Pu-240

As noted earlier, some of the neutron absorptions in the Pu-239 (about 27%) produce Pu-240 which is non-fissile, but has a fairly large absorption cross-section for thermal neutrons (290 barns). As significant amounts of Pu-239 begin to build up in the fuel, Pu-240 will also start to appear. Although in principle Pu-240 also eventually reaches an equilibrium value, actual fuel irradiation times are much too short for this to occur, and as far as we are concerned, Pu-240 is a poison that increases at a steady rate as the fuel burns up. Pu-240 does however have one useful characteristic: neutron capture in Pu-240 creates Pu-241, a fissile material similar in properties to Pu-239. This provides only minor compensation for overall reactivity loss because only small quantities of Pu-241 can be produced over the typical fuel irradiation time.

7.4.3 Changes in Reactivity with Burnup

Figure 7.3 shows the concentrations of the fissile isotopes U-235, Pu-239 and Pu-241 as a function of total fuel exposure. The same data are presented in Table 7.1, in terms of both burnup units. (The two units are not strictly linearly related because the ratio of energy extracted from the fuel to its neutron exposure varies slightly with the changing composition of the fuel).

In the figure, the approximate exit irradiation for Point Lepreau, Pickering and Bruce are indicated. The higher burnup achieved by the Bruce A reactor is due to the fact that it does not use adjuster rods for flux flattening, and therefore does not suffer the associated reactivity penalty. There is, however, a large difference in fuel burnup in the outer zone of Bruce compared with the inner, since a high degree of differential burnup is used in place of adjusters for flux flattening.

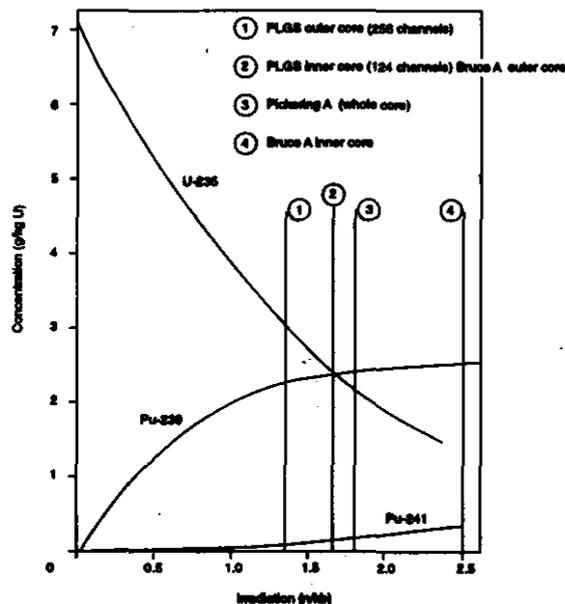


Figure 7.3: Fissile isotope concentrations as a function of burnup

Fissile isotope concentrations as a function of burnup

Table 7.1
Burnup Data*

n/kb	MWh/kgU	U-235 (g/kgU)	Pu-239 (g/kgU)	Pu-241 (g/kgU)
0	0	7.20	0	0
0.2	19	6.37	0.60	0.002
0.4	39	5.62	1.10	0.009
0.6	59	4.90	1.48	0.025
0.8	79	4.30	1.77	0.049
1.0	100	3.76	1.98	0.078
1.2	120	3.32	2.14	0.107
1.4	140	2.90	2.25	0.145
1.6	159	2.56	2.33	0.177
1.8	179	2.26	2.39	0.211
2.0	198	1.98	2.43	0.245
2.2	216	1.74	2.46	0.278
2.4	235	1.54	2.48	0.309
2.6	253	1.35	2.49	0.338
2.8	271	1.18	2.50	0.366
3.0	289	1.03	2.50	0.393

* Strictly speaking, the values shown in this table apply only to the Pickering reactors, but they will be correct to within a percent or so for all natural uranium, D₂O moderated reactors.

The more important components of the change of reactivity with irradiation are shown in Figure 7.4. Initially, as indicated by the combined curve (U-235 + Pu-239), the positive reactivity contribution of the Pu-239 overcomes the negative contribution due to burnup of U-235, so that the net effect is an increase of reactivity. Although only eight Pu-239 atoms are produced for every ten U-235 destroyed, the higher fission cross-section of the Pu-239 (742 b compared to 580 b for U-235) more than compensates for this ratio being less than one. Eventually, the Pu-239 buildup rate falls off as it moves towards equilibrium (although its stay in the reactor is not long enough for it to get very close to the equilibrium value). The fall-off in Pu-239 production means that it can no longer compensate for the continuing burnup of the U-235 and the curve turns over.

Reactivity change with burnup

The creation of Pu-240 produces the steady reactivity decrease shown in the figure, partially compensated by the buildup of fissile Pu-241. The neutron-absorbing fission products cause a negative reactivity which builds up continuously as irradiation proceeds. Note that the effect of the very strongly absorbing fission product xenon-135 is *not* included in Figure 7.4. Xenon is so important, and it affects reactivity so dramatically in the short-term that this will be covered in detail in Module 11.

The rate at which any given fission product builds up depends on the magnitude of its absorption cross-section. Through an argument similar to the one in Section 7.4.2 for Pu-239, we can see that each fission product will approach its equilibrium concentration at a rate which depends on its absorption cross-section (equation 7.6). The high cross-section fission products will approach equilibrium relatively quickly; the initial steep part of the fission product reactivity curve in Figure 7.4 is due to these, and particularly to samarium-149 (absorption cross-section 42,000 b), which will also be considered in detail in Module 11. The gradual longer-term variation of the fission product reactivity is due to the buildup of the weaker absorbers.

The net effect of all these contributions is shown by the “total” curve in Figure 7.4. The initial drop (lasting a few days) is due to the predominant effect of the buildup of Sm-149. Once this reaches equilibrium, the net positive contribution from the U-235 and Pu-239 keeps the reactivity change positive up to an exposure of about 1 n/kb. Beyond that point, reactivity decreases as U-235 continues to burn up, the Pu-239 growth rate falls off as it moves toward equilibrium, and the fission products and Pu-240 steadily build up in the fuel. The Pu-241 reduces that rate of reactivity loss, but is unable to reverse it. At some point, therefore, it will be necessary to start removing partially burnt-up fuel, replacing it with fresh fuel. The practical aspects of refuelling will be considered in Section 7.6.

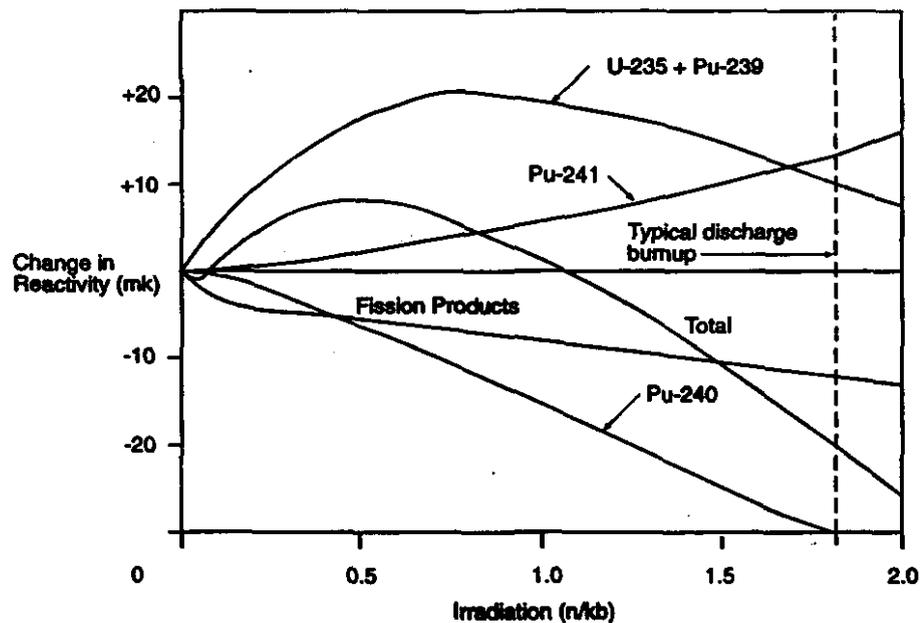


Figure 7.4: Components of Change of Reactivity with Burnup

Another way of seeing what happens in a fuel bundle as a result of neutron exposure is to see how the k_{∞} value of the fuel changes as it burns up. The variation of k_{∞} , and the four factors contributing to it, is illustrated in Figure 7.5. We should first note that there is no detectable change in either the fast fission factor (ϵ) or the resonance escape probability (p). This is to be expected, because these factors are determined by the quantity of U-238 in the fuel, and the percentage change of the U-238 is very small (it constitutes about 99% of the fuel regardless of whether it has been irradiated or not).

k_{∞} as a function of burnup

The most important change concerns the reproduction factor (η). We recall from Section 5.4 that:

$$\eta = \nu \frac{\Sigma_f(\text{fuel})}{\Sigma_a(\text{fuel})}$$

The sharp initial dip in η comes from the accumulation of the fission product samarium-149, which builds up to its equilibrium value over the first few days of operation. Since the term "fuel" is defined to include all materials in the fuel matrix, Sm-149, with its high thermal cross-section of 4.2×10^4 barns, increases the absorption cross-section of the fuel bundle significantly without affecting its fission cross-section. The value of η then gradually increases because the initial buildup of Pu-239 more than compensates for the burnup of U-235. After reaching a peak, it declines again due to the continued burnup of U-235, a slowing of the growth of Pu-239 and a buildup of Pu-240 and fission products. (Note again that the effect of xenon-135 is *not* included in the figure.)

The thermal utilization (f) shows a slight increase due to the increasing absorption in the fuel relative to the core structural materials. (Note that the buildup of Pu-239, Pu-240, Pu-241 and the fission products all lead to an increased absorption by the fuel).

The variation of k_{∞} and k is also shown in Figure 7.5. The shape of both resembles that of the η -variation. Since irradiation does not alter non-leakage factors much, the curve for k is almost identical in form to that of k_{∞} , but is displaced downwards by about 30 mk.

Since the long-term effect of irradiation on k is a gradual decrease, the reactor eventually runs out of reactivity and we have to start replacing the fuel. Normally we target our reactors to operate at full power with small amounts of positive reactivity (typically about 5 mk) available in addition to the xenon override capability. Figure 7.6 shows the "excess reactivity" above this target level (the excess reactivity is held down in practice by having the appropriate quantity of poison in the moderator).

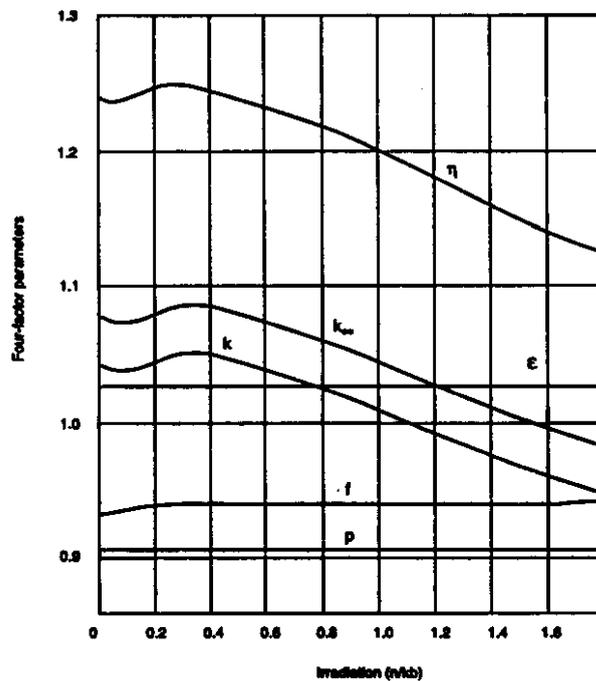


Figure 7.5: Variation of four-factor parameters, k_{∞} and k with burnup

The figure shows that to maintain target reactivity, refuelling was started after about 180 full-power days. From this point on, the reactor is in an *equilibrium* fuel condition (we refer to its state before this as the *fresh* fuel condition). Fuel is now replaced on a daily basis (between 8 and 18 bundles per day) to add reactivity at a rate equal to its rate of loss from burnup. The average burnup of fuel discharged from the reactor is larger than one might expect from the previous graph (Figure 7.5), because that refers to a core which is assumed to be burning up uniformly, while the actual reactor contains a mixture of fuel bundles of differing degrees of irradiation. Some of these bundles (the ones nearing discharge) will have an irradiation appreciably greater than that permitted by the "uniform core" calculation, but the reactivity "deficit" they cause is compensated by the bundles whose irradiation is below average.

Equilibrium fuel

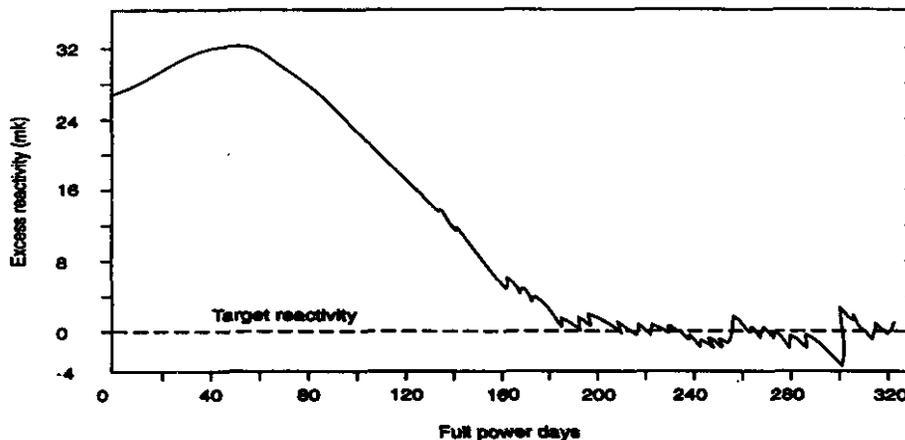


Figure 7.6: Reactivity as a function of burnup

Change in β with burnup

7.5 EFFECT OF BURNUP ON REACTOR - KINETICS

The main effect on reactor kinetics is the change in the overall delayed neutron fraction as U-235 is burnt up and replaced by Pu-239. As mentioned in Section 2.8.5, the delayed neutron fraction (β) for U-235 is 0.0070, while that for Pu-239 is only 0.0023. As the fuel composition changes, therefore, there will be a gradual reduction in the effective value of the delayed neutron fraction. This will have a direct effect on how quickly the reactor responds to any changes in reactivity. This point will be considered in detail in Module 8.

7.6 FUEL MANAGEMENT - PRACTICAL ASPECTS

The Fuel Engineer at the station is responsible for ensuring, as much as possible, that the optimum fuel cycle is used. The overall objective is to maintain maximum reactor power output at minimal fuel cost, while avoiding any over-rating of the fuel or any significant asymmetries in the power distribution.

Various computer programs exist which can follow the histories of bundles in the core. For example, these programs calculate the expected axial and radial power distributions, the burnup of each bundle in the core and the excess reactivity available. The validity of these calculations can be checked by comparing the power distributions put out by the program with those obtained from the flow rates and temperature increases (ΔT) in various channels. If there are large discrepancies, the physics data of the program are modified by intelligent guesses until eventually the agreement between theory and practice is close enough.

The Fuel Engineer uses the output from this type of program to help decide which channels to fuel and when. Since the core is usually divided into a number of annular zones of roughly equal ratings, the fuelling rates per zone can easily be derived. Even so, no rigid fuelling pattern is used. The following criteria would have to be considered:

1. discharge of highest burnup fuel (this information is obtained from the program)
2. high reactivity gain per channel fuelling
3. no fuelling in high power areas if derating is likely to be necessary (the fuelling program will print out a power matrix)
4. preservation of symmetry
5. equal numbers fuelled per liquid control zone
6. fuelling from alternate ends of reactor
7. effect on neighbouring channels
8. special requirements of experimental bundles
9. priority to channels known to contain failed fuel
10. maintain margin to trip

Once a channel has been fuelled, the corresponding changes in bundle positions will have to be entered for the next run of the computer program. If the axial flux distribution in the reactor is fairly flat, it might be expedient to fuel in so-called 8 or 10 bundle shifts. Figure 7.7 shows the changes in bundle positions for an 8-bundle shift. The advantages of the move in equalizing the total neutron exposure of the outermost bundles are obvious.

Fuelling criteria

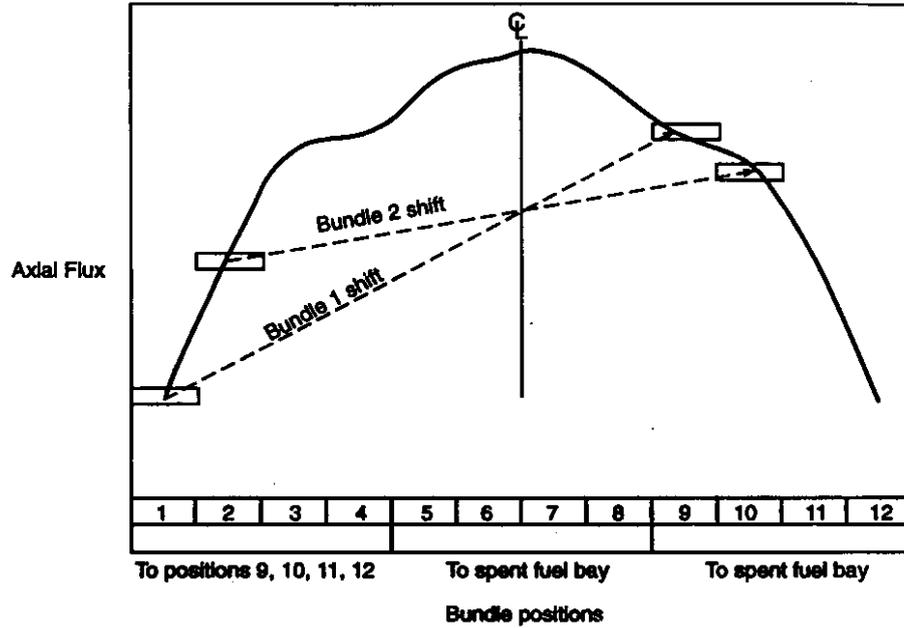


Figure 7.7: Changes in bundle positions for 8-bundle shift

ASSIGNMENT

1. Write the equations for the rate of destruction of U-235 (dN_5/dt) and the rate of build-up of Pu-239 (dN_9/dt), and sketch the way in which N_5 and N_9 vary with time in a reactor.
2. Using equation 7.2, calculate the time required for the U-235 concentration to be reduced to one half of its start-of-life value (assume that the average thermal neutron flux in the fuel is 10^{14} n/cm²s).
3. Using equation 7.6, calculate the time required for Pu-239 to build up to one half of its equilibrium concentration (assume that the average thermal neutron flux in the fuel is 10^{14} n/cm²s).
4. Explain how and why the reproduction factor (η) changes from fresh to equilibrium fuel in a CANDU reactor.
5. Using Figure 7.3, calculate the total fissile content of the fuel at exit from a Pickering reactor, as a percentage of the initial fissile content. Inasmuch as the percentage you have calculated is still rather high, explain why the fuel isn't left in the reactor longer.
6. Using equation 7.5, calculate the ratio of the equilibrium Pu-239 concentration to the initial concentration of U-235 in the fuel.