Core Composition Changes

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Summary:
Fission product poisoning and fuel depletion are investigated. Models are proposed and analytical solutions are obtained for a few simple, but illustrative, cases.

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Figure 1 Course Map.

1.2 Learning Outcomes

The goal of this chapter is for the student to understand:

- The phenomenon of fission product poisoning, how to model it and how it effects fuel cross section and reactivity.
- The phenomenon of fuel depletion, how to model it and how it effects fuel cross section.

2 Fission Product Poisoning

There are two major dynamic phenomena that affect the fuel composition: fission product poisoning (with a time scale of hours and days) and fuel depletion (with a time scale of days and months). In this section we will consider fission product poisoning.

There is a full spectrum of isotopes formed when a fission occurs. Two of the many isotopes produced are of particular concern, xenon-135 (Xe-135) and samarium 149 (Sm-149) because they have high absorption cross sections and, so, act as parasites or poisons which tend to shut the reactor down unless compensated for. Xe-135 has a cross section of about 10^6 barns while Sm-149 has a cross section of about 10^5 barns. These are orders of magnitude higher than the typical isotopes found in a reactor, so even small quantifies have a noticeable affect.

Apparently, this affect was not anticipated when the first reactor was started up, or so the story goes. The scientists of the day felt confident in their measurements and predicted that the core would be of a certain size when it when critical. The engineers in the crowd felt that extra allowances should be made to account for the unforeseen. When the reactor when critical as the scientists predicted, they felt vindicated, only to be proven wrong when the reactor shut itself down after a short operation. The engineer's prudence proved correct in the end. True or not, it makes for a good story and motivation for this chapter.

2.1 Effect on Reactivity

To estimate the magnitude of the effect of fission product poisoning on reactivity, we note that

$$\rho = \text{reactivity} \equiv \frac{k-1}{k} \tag{2.1}$$

We will focus on the infinite reactor, ie, we will ignore leakage effects and deal only with k_∞:

$$k_\infty =, \ \eta f p \tag{2.2}$$

The factor f, the thermal utilization, is the factor most affected by the addition of poisons in the fuel:

$$f' = \frac{\Sigma_a^{fuel}}{\Sigma_a^{fuel} + \Sigma_a^{moderator} + \Sigma_a^{poison}} \tag{2.3}$$

So, comparing the 'before' and 'after' poison cases:

$$\Delta\rho \equiv \rho' - \rho = \frac{k'-1}{k'} - \frac{k-1}{k} = \frac{\cancel{kk'}-k-\cancel{k'k}+k'}{k'k} = \frac{k'-k}{k'k} = \frac{f'-f}{f'k} \simeq \frac{f'-f}{f'} \text{ since } k \simeq 1$$

$$= \frac{\dfrac{\sum_a^{fuel}}{\sum_a^{fuel}+\sum_a^{mod\,erator}+\sum_a^{poison}} - \dfrac{\sum_a^{fuel}}{\sum_a^{fuel}+\sum_a^{mod\,erator}}}{\dfrac{\sum_a^{fuel}}{\sum_a^{fuel}+\sum_a^{mod\,erator}+\sum_a^{poison}}} \qquad (2.4)$$

$$= 1 - \frac{\sum_a^{fuel}+\sum_a^{mod\,erator}+\sum_a^{poison}}{\sum_a^{fuel}+\sum_a^{mod\,erator}} = -\frac{\sum_a^{poison}}{\sum_a^{fuel}+\sum_a^{mod\,erator}}$$

So, the negative reactivity insertion is proportional to the amount of poison. This is no surprise.

2.2 Production and Decay

Xenon-135 is the most important poison. It is produced directly as a fission product but it is also produced indirectly by the decay of antimony-135, another fission product. Looking at the decay scheme depicted by figure 2, we see that antimony-135 decays in a matter of seconds to iodine-135 and then to xenon-135 with a half-life of 6.58 hours. The xenon then decays more slowly with a half-life of 9.17 hours so that it tends to accumulate. But because Xe-135 has a high absorption cross section, it has a tendency to absorb neutrons and be transformed into another isotope that has a low cross section. In this manner, Xe-135 is removed and is no longer around to act as a poison. Obviously, we need to quantify the ebb and flow of Xe-135 levels is we are to be able to accommodate and counteract poison effects on criticality.
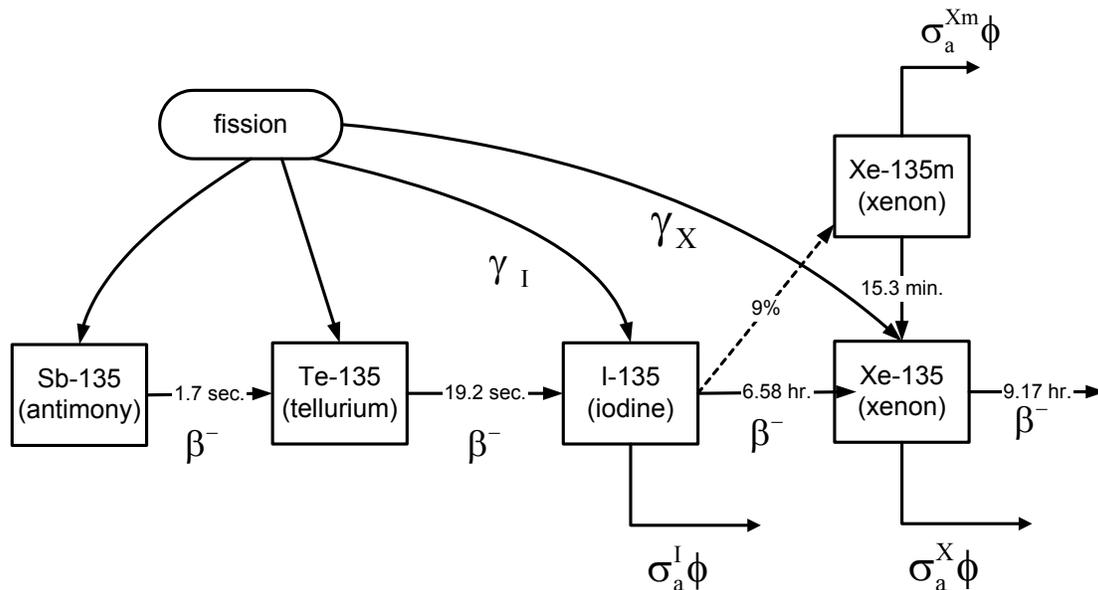
Figure 2 Iodine and xenon production and loss scheme.

The governing equations are:

$$\frac{\partial I(\mathbf{r},t)}{\partial t} = \underbrace{\gamma_I \Sigma_f \, \phi(\mathbf{r},t)}_{\text{fission production}} - \underbrace{\lambda_I I(\mathbf{r},t)}_{\text{decay}}$$

$$\frac{\partial X(\mathbf{r},t)}{\partial t} = \underbrace{\gamma_X \Sigma_f \, \phi(\mathbf{r},t)}_{\text{fission production}} + \underbrace{\lambda_I I(\mathbf{r},t)}_{\text{decay source}} - \underbrace{\lambda_X X(\mathbf{r},t)}_{\text{decay}} - \underbrace{\sigma_a^X \phi(\mathbf{r},t) X(\mathbf{r},t)}_{\text{absorption loss}}$$

(2.5)

Note that it is assumed that the fission products do not migrate in space, ie that they stay where they are produced. This is typically valid for solid fuels where migration is limited to some local movement through the fuel grain structure or minor ceramic fuel cracks. The fission product densities are space dependent, however, since the flux is space dependent. To solve these equations, we need to specify the initial conditions. We look at two illustrative cases: startup and shutdown.

2.3   Clean Core Startup

We ignore the special dependence of the flux. Any solution we find will be applicable for that flux value.

Initial conditions of constant flux with a clean core (fresh fuel, no poisons):

$$X(0) = 0 = I(0)$$

$$\phi(t) = \phi(0) = \text{constant}$$

(2.6)

The solution for the iodine history is:

$$I(t) = \underbrace{\frac{\gamma_I \Sigma_f \, \phi_0}{\lambda_I}}_{I(\infty)} \left(1 - e^{-\lambda_I t}\right)$$

(2.7)
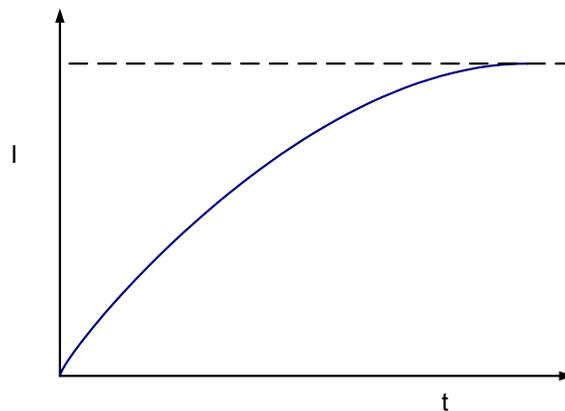
Figure 3 Iodine level after startup

The xenon solution is more complex but still can be obtained analytically:

$$X(t) = \underbrace{\frac{(\gamma_I + \gamma_X)\Sigma_f\,\phi_0}{\lambda_X + \sigma_a^X\phi_0}}_{X(\infty)}\left[1 - e^{-(\lambda_X + \sigma_a^X\phi_0)t}\right] + \frac{\gamma_I\Sigma_f\,\phi_0}{\lambda_X - \lambda_I + \sigma_a^X\phi_0}\left[e^{-(\lambda_X + \sigma_a^X\phi_0)t} - e^{-\lambda_I t}\right] \qquad (2.8)$$

Knowing X(t), we can calculate the poison cross section:

$$\Sigma_a^{poison}(t) = \Sigma_a^X(t) = \sigma_a^X X(t)$$

$$\Rightarrow \Delta\rho = -\frac{\Sigma_a^{poison}(t)}{\Sigma_a^{fuel} + \Sigma_a^{moderator}} = -\frac{\sigma_a^X X(t)}{\Sigma_a^{fuel} + \Sigma_a^{moderator}} \qquad (2.9)$$

The long term, or saturation, value of xenon is $X(\infty)$ as defined in equation 2.8. Note that the saturation value is **not** a linear function of the flux, while the saturation value for iodine is. To illustrate this, Xe vs ϕ is plotted in figure 4. **Note that this is not Xe vs time.**
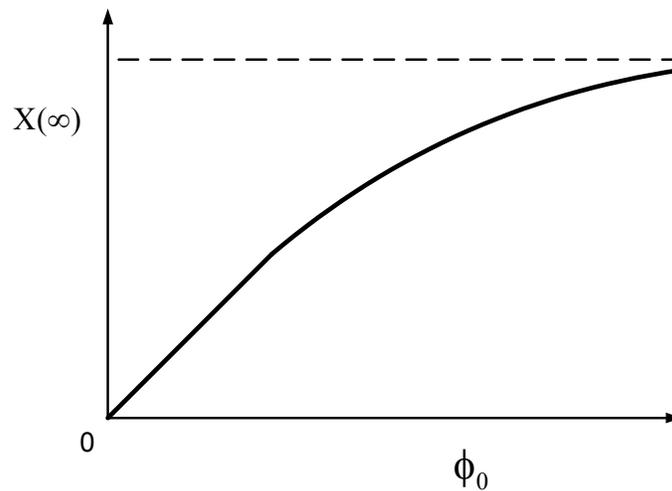
Figure 4 Saturation xenon level vs flux.

At low flux levels, the saturation xenon level is a linear function of flux since there is insufficient flux to burn off the existing xenon. At sufficiently high flux levels, however, a balance is reached where xenon production is offset by the absorption losses. Flux levels found in typical power reactors and higher power research reactors lie about in the middle of the graph.

2.4 Shutdown

We start from the initial condition that the reactor has been operating for a long time. Then we shut the reactor down. We assume that the flux is zero after shutdown. Thus we have:

$$X(0) = X(\infty)$$

$$I(0) = I(\infty) \qquad (2.10)$$

$$\phi(t) = \phi(0) = 0$$

The governing equations are the same as previously except that all the terms involving the flux are zero, ie there is no fission source and there is no absorption sink:

$$\frac{\partial I(\mathbf{r},t)}{\partial t} = -\underbrace{\lambda_I I(\mathbf{r},t)}_{\text{decay}}$$

$$\frac{\partial X(\mathbf{r},t)}{\partial t} = +\underbrace{\lambda_I I(\mathbf{r},t)}_{\text{decay source}} - \underbrace{\lambda_X X(\mathbf{r},t)}_{\text{decay}}$$

(2.11)

Solving, again ignoring the spatial dependence, we get

$$I(t) = I_\infty e^{-\lambda_I t}$$

$$X(t) = X_\infty e^{-\lambda_X t} + \underbrace{\frac{\lambda_I I_\infty}{(\lambda_I - \lambda_X)}\left(e^{-\lambda_X t} - e^{-\lambda_I t}\right)}_{\text{this term is} > 0}$$

(2.12)

Now, since

$$\left(T_{1/2}^I = 6.58\text{hr}\right) < \left(T_{1/2}^X = 9.17\text{hr}\right)$$

(2.13)

we have

$$\lambda_I > \lambda_X$$

(2.14)

thus the final term on the RHS of equation 2.12 is positive and hence causes the Xe level to rise immediately after shutdown. The Xe level is something other than a simple decay, as shown in figure 5, because the decaying iodine provides a source for Xe. Immediately before the reactor shutdown, the Xe level is the saturated value, $X(\infty)$. Since all terms have exponentials that decay, the Xe level eventually decays away. The height of the peak depends on the values of $X(\infty)$ and $I(\infty)$ which depend on ϕ_0.
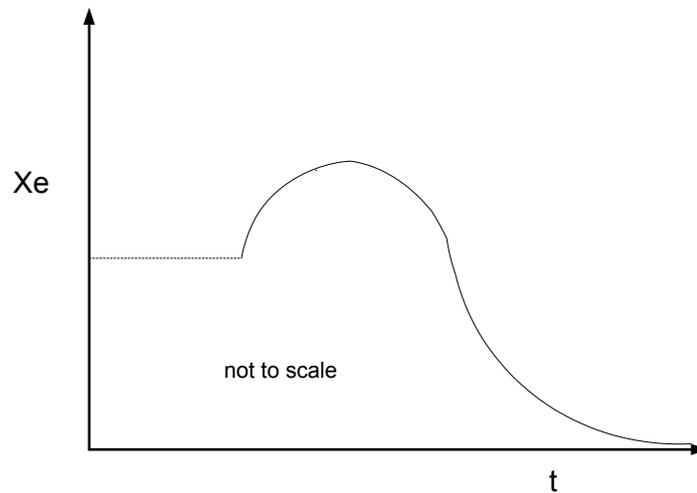
Figure 5 Xe vs time after shutdown.

2.5 Poison-out

Because the Xe builds-in after shutdown, negative reactivity is inserted and control absorbers will have to be removed from the reactor to compensate is criticality is to be maintained. The typical reactor only has a limited excess reactivity so that an excessive amount of positive

reactivity cannot be inserted accidentally and because it is costly to deliberately run a reactor with a large amount of absorbers inserted into the core. In the CANDU reactor, there is only enough excess reactivity to compensate for about 20 to 30 minutes of Xe buildup. Thus the operator has 2-30 minutes to restart the reactor after a reactor trip. Beyond that, the operator will have to wait about 40 hours for the Xe to decay sufficiently to be able to start up the reactor, as illustrated in figure 6.
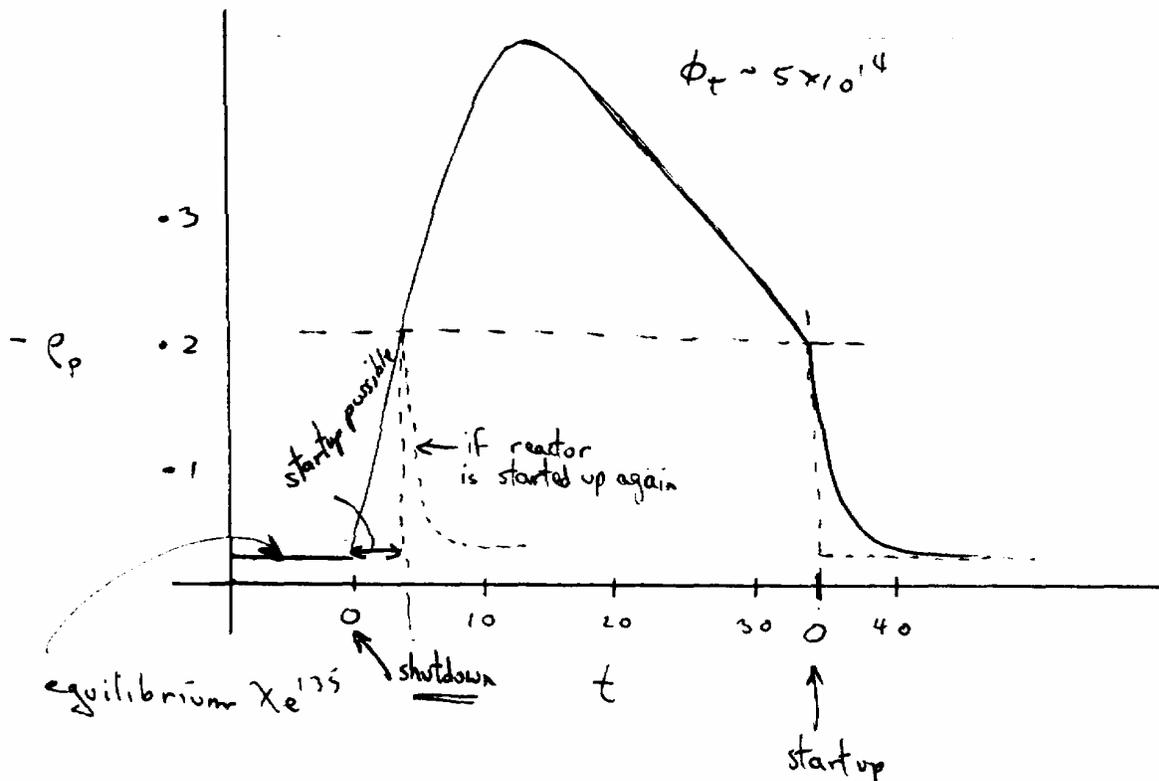
Figure 6 Xenon buildup and startup capability.

If the shutdown is planned or if the operator has some warning, the reactor can be operated at a lower power (say 60% or some lower power that is high enough to prevent poison out) until a new equilibrium is reached. At a lower power, the equilibrium amount of Xe, and hence the peak reactivity insertion, is lower. Thus it would be easier to restart from this lower power. See figure 7.
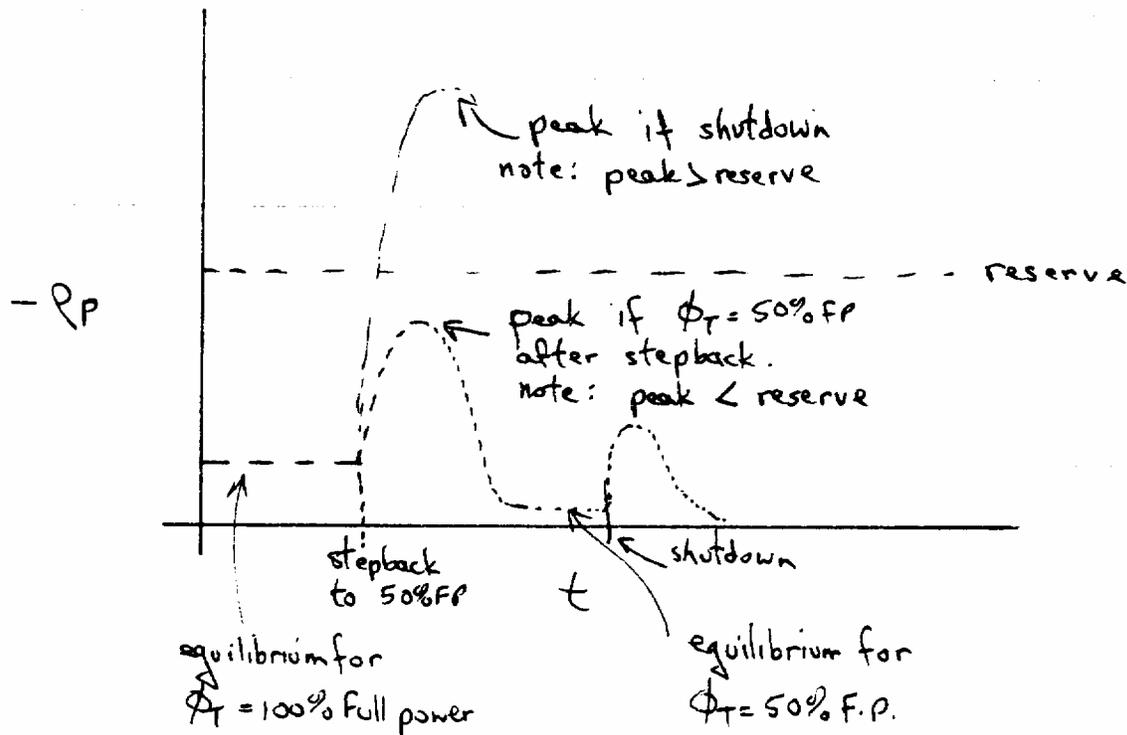
Figure 7 Startup strategy.

With the revised shutdown scheme, the operator can startup at any time, for example, when minor repairs have been completed, rather than be forced to wait 40 hours at ~$10,000/hr for replacement electricity from a coal-fired station.

2.6 Xenon Oscillation

In a finite reactor with a space dependent flux and material properties, with space dependent burnup and control rod action, a local region may see an increased flux for a period of time. This causes an increased Xe burnout and hence a lower ρ_p. With this increase in local reactivity, the local flux increases still further. The overall control scheme will insert control rods globally to compensate and cause a flux depression elsewhere. The region of depressed flux sees the opposite effect, ie a local buildup of Xe. These effects are limited by the increased production of Xe via I for the high flux region and the opposite for the low flux region. The time delays inherent in the process leads to a high ρ_p in the high flux region due to the higher production. Hence, ρ_p eventually increases causing the flux to depress. Now this region becomes the low flux region. The period of the oscillations is ~15-20 hours and occurs in large reactors like the CANDU and where the flux is $> \sim 10^{13}$ n /cm^2 – s. The large size of the reactor, compared to the neutron diffusion length, allows regions of the core to behave pseudo-independently and the high flux gives significant Xe burnup, both of which are necessary for the oscillations to occur.

These oscillations can be important since they may lead to local flux, power and temperature conditions which are unacceptable from a materials point of view and hence lead to safety problems.

2.7 Permanent Poisons

In addition to xenon, other fission products, like samarium, have a high enough cross section to be be an effective neutron absorber. But the cross section is not high enough to be burned off by the flux and does not decay. Hence, the poison grows in and stay in the fuel. Thus the governing equation is:

$$\frac{\partial N_{PP}(\mathbf{r},t)}{\partial t} = \underbrace{\gamma_{PP} \Sigma_f(\mathbf{r},t)\phi(\mathbf{r},t)}_{\text{fission production}} = \gamma_{PP} \Sigma_f(\mathbf{r},0)\phi(\mathbf{r},0) = \gamma_{PP} \cdot \text{ constant} \tag{2.15}$$

which has as a solution:

$$N_{PP}(\mathbf{r},t) = \gamma_{PP} \Sigma_f(\mathbf{r},0)\phi(\mathbf{r},0)t = \gamma_{PP} \cdot \text{ constant} \cdot t \tag{2.16}$$

assuming we start from fresh fuel. The permanent poison cross section is thus:

$$\Sigma_a^{PP}(\mathbf{r},t) = \gamma_{PP}\sigma_a^{PP} \Sigma_f(\mathbf{r},0)\phi(\mathbf{r},0)t = \text{constant} \cdot t \tag{2.17}$$

3 Fuel Depletion

As the fission process occurs, the fuel is slowly depleted. Given the fuel density (about 10^{22} nuclei / cm^3) compared to the neutron flux density (about 10^{14} n / $cm^2 - s$), it takes days to months to see appreciable fuel depletion. It is reasonable, then, to solve the fuel isotopic balance equations as a function of time for a relatively stable flux. We solve for the steady state flux for a given fuel composition. This gives the flux to use in the depletion equations. When the fuel composition changes significantly, we update the steady state flux, and so on. We keep the flux steady by control rod action.

3.1 Simple Depletion Dynamics

In general, the number of isotopes to track can be large. Computer codes such as the SCALE package are typically used in the industrial setting. Herein, we will just look at a few simple cases to illustrate some key features.

For 1 fuel isotope:

$$\frac{\partial N_f}{\partial t} = -N_f(\mathbf{r},t)\sigma_a^{fuel}\phi(\mathbf{r},t) \tag{3.1}$$

which has as a general solution for time varying flux:

$$N_f(\mathbf{r},t) = N_f(\mathbf{r},0)e^{-\sigma_a^{fuel}\int_0^t \phi(\mathbf{r},t')dt'} \tag{3.2}$$

The integrated flux occurs sufficiently often to be given a special name: **neutron fluence**:

$$\Phi(\mathbf{r},t) = \int_0^t \phi(\mathbf{r},t')dt' \equiv \text{neutron fluence} \tag{3.3}$$

Of course, equation 3.3 cannot be solved unless we know the flux. But the flux depends on the isotopic composition of the fuel, ie N_f! Normally we get around this issue by solving the full equation set numerically. This is not hard to do. Just use a simple Euler solver that we have seen before. To get analytical solutions, we pose two special cases.

The first case is $\phi = $ constant. Equation 3.2 becomes:

$$N_f(\mathbf{r},t) = N_f(\mathbf{r},0)e^{-\sigma_a^f \phi_0(\mathbf{r})t} \tag{3.4}$$

Figure 8 illustrates the exponential burnup.

Case 2 is power = constant. Now, power density is just the fission rate x energy released per fission (w_a):

$$P(\mathbf{r},t) = w_a \Sigma_a^{fuel}(\mathbf{r},t)\phi(\mathbf{r},t) = w_a N_f(\mathbf{r},t)\sigma_a^{fuel}\phi(\mathbf{r},t) = P_0(\mathbf{r}) \tag{3.5}$$

Since Nf is decreasing with time, the flux will be increasing to compensate. Thus equation 3.1 becomes:

$$\frac{\partial N_f(\mathbf{r},t)}{\partial t} = -N_f(\mathbf{r},t)\sigma_a^{fuel}\phi(\mathbf{r},t) \Rightarrow -\frac{P_0(\mathbf{r})}{w_a} \tag{3.6}$$

which has the simple solution:

$$N_f(\mathbf{r},t) = N_0(\mathbf{r},0) - \frac{P_0(\mathbf{r})}{w_a}t \tag{3.7}$$

Both approximations (constant flux and constant power) are valid for short times and give the same answer in the limit as $t \to 0$, as suggested in figure 8.
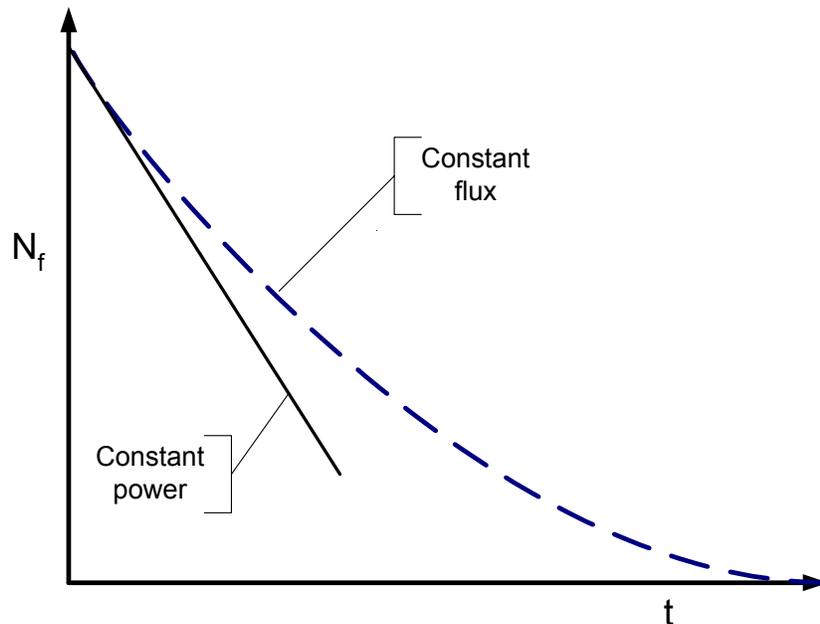
Figure 8 Fuel depletion over time.

3.2 Simple Reactor Model

Now that we have seen how to calculate how the fuel depletes over time, let's look at how k_∞ is affected. Assume an infinite, critical, homogeneous reactor:

$$k_\infty =, \ \eta f p =, \ \eta p \frac{\Sigma_a^{fuel}(t)}{\Sigma_a^{fuel}(t) + \Sigma_a^{moderator} + \Sigma_a^{poison}(t) + \Sigma_a^{control}(t)} \tag{3.8}$$

As the poison builds-in and the fuel depletes, $\Sigma_a^{fuel}(t) \to 0$, $\Sigma_a^{poison}(t) \uparrow$, and $\Sigma_a^{control}(t) \to 0$, that is, the control rods are withdrawn to compensate for the loss of fuel and the addition of poison.

For the constant power case, we had:

$$\begin{aligned} N_f(\mathbf{r}, t) &= N_f(\mathbf{r}, 0) - N_f(\mathbf{r}, 0)\sigma_a^{fuel}\phi(\mathbf{r}, 0)t \\ &= N_f(\mathbf{r}, 0)\left(1 - \sigma_a^{fuel}\phi(\mathbf{r}, 0)t\right) \end{aligned} \tag{3.9}$$

Multiplying both sides by σ_a^{fuel}

$$\sigma_a^{fuel}N_f(\mathbf{r}, t) = \sigma_a^{fuel}N_f(\mathbf{r}, 0)\left(1 - \sigma_a^{fuel}\phi(\mathbf{r}, 0)t\right)$$
$$\Downarrow \tag{3.10}$$
$$\Sigma_a^{fuel}(\mathbf{r}, t) = \Sigma_a^{fuel}(\mathbf{r}, 0)\left(1 - \sigma_a^{fuel}\phi(\mathbf{r}, 0)t\right)$$

Thus we have the fuel cross section to use in equation 3.8.

We need the poison history as well. From equation 2.8 we found that

$$X(\infty) = \frac{(\gamma_I + \gamma_X)\Sigma_f \phi_0}{\lambda_X - \sigma_a^X \phi_0} \tag{3.11}$$

But the flux has to increase as the fuel is depleted to keep the power constant. Thus:

$$\Sigma_f \phi_0 = \text{constant, but } \phi_0 \uparrow \text{ as } t \uparrow \tag{3.12}$$

We get the flux variation in time from the constant power assumption:

$$P(\mathbf{r}, t) = w_a \Sigma_a^{fuel}(\mathbf{r}, t)\phi(\mathbf{r}, t) = w_a \Sigma_a^{fuel}(\mathbf{r}, 0)\phi(\mathbf{r}, 0) = P_0(\mathbf{r})$$
$$\Downarrow \tag{3.13}$$
$$\phi(\mathbf{r}, t) = \frac{\Sigma_a^{fuel}(\mathbf{r}, 0)\phi(\mathbf{r}, 0)}{\Sigma_a^f(\mathbf{r}, t)} = \frac{\Sigma_a^{fuel}(\mathbf{r}, 0)\phi(\mathbf{r}, 0)}{\Sigma_a^{fuel}(\mathbf{r}, 0)\left(1 - \sigma_a^{fuel}\phi(\mathbf{r}, 0)t\right)} = \frac{\phi(\mathbf{r}, 0)}{\left(1 - \sigma_a^f\phi(\mathbf{r}, 0)t\right)}$$

This flux is the flux to use in equation 3.12. Thus:

$$\Sigma_a^X(\mathbf{r}, t) = \sigma_a^X X(\infty) = \frac{(\gamma_I + \gamma_X)\Sigma_f^{fuel}(\mathbf{r}, 0)\phi(\mathbf{r}, 0)}{\dfrac{\lambda_X}{\sigma_a^X} - \phi(\mathbf{r}, t)} \tag{3.14}$$

where $\Sigma_f^{fuel}(\mathbf{r}, 0)\phi(\mathbf{r}, 0)$ is a constant since the fission cross section for fuel is changing in proportion to the fuel absorption cross section.

We determined the permanent poison cross section in equation 2.17 so now we have all we need for the determination of k_∞ via equation 3.8. To get an estimate of how long it would take before the control rods are completely withdrawn in the attempt to compensate for fuel depletion and poison build-up, we set $\sum_a^{control}(t) = 0$ and solve equation 3.8 for time, t. This would represent some upper limit on the core loading lifetime. Once there are no absorbers left to remove, refueling would have to occur.

In CANDU reactors, which are refueled on-line, this does not represent an operational restriction. Other power reactors, however, are batch refueled and outages are required to refuel. This is usually carefully planned for and performed during the yearly maintenance outage. The requirement for having extra absorbers in the core to compensate for a year's burnup exacts a large fuel cost penalty in such reactors. Damaged fuel cannot be readily removed in a batch fuelled reactor until the scheduled refueling time whereas in CANDU reactors, it can be removed soon after detection. This reduces the fission product release to the heat transport system in CANDU reactors and is one advantage of CANDU over other reactor types.

3.3   General Depletion Dynamics

The general isotope history can be followed by modeling all the processes involved:
1. loss by decay of the isotope
2. loss by neutron capture by the isotope which transmutes it into another isotope of the same element
3. production by decay from a parent isotope
4. production by capture transmutation
5. imposed fuel loading changes.

These 5 processes are illustrated in figure 9.
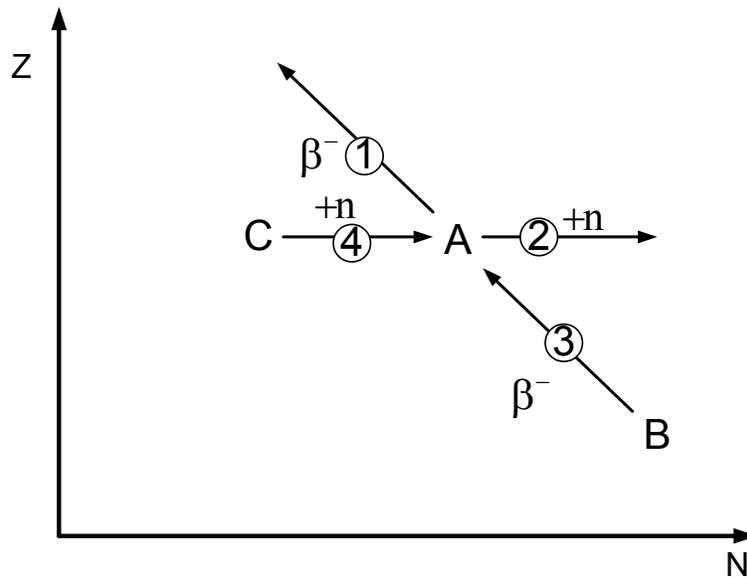
Figure 9 Isotope transformation processes.

The governing rate equation is:

$$\frac{dN_A}{dt} = \underbrace{-\lambda_A N_A}_{\text{decay}} \underbrace{-\left[\sigma_a^A \phi\right] N_A}_{\text{neutron capture}} + \underbrace{\lambda_B N_B}_{\text{parent decay}} + \underbrace{\left[\sigma_\gamma^C \phi\right] N_C}_{\text{capture transmutation}} + \underbrace{F(t)}_{\text{fuel loading}} \qquad (3.15)$$

We can safety assume that the flux is constant over the integration time-step and easily solve this equation numerically.

In general, there are many isotopes to track so we solve them all simultaneously.

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