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Nuclear Theory - Course 227

FISSION PRODUCT POISONING

All Fission products can be classified as reactor poisons because they all absorb neutrons to some extent. Most simply buildup slowly as the fuel burns up and are accounted for as a long term reactivity effect (as we did in lesson 227.00-7). However, two of the fission products, Xe-135 and Sm-149, are significant by themselves due to their absorption cross section and high production as fission products or fission product daughters. Xenon-135 has a microscopic absorption cross section of 3.5×10^6 barns and a total fission product yield of 6.6%. Samarium-149 has an absorption cross section of 42,000 barns and a total fission product yield of 1.4%. Xenon-135 is the more important of the two and will be dealt with in more detail.

Xenon-135

Xenon-135 (often carelessly referred to just as xenon) is produced in the fuel in two ways:

- a) Directly from fission. About 0.3% of all fission products are Xe-135.
- b) Indirectly from the decay of iodine-135, which is either produced as a fission product or from the decay of the fission product tellurium-135 via the following decay chain:

 $_{52}Te^{135} \xrightarrow{\beta, \gamma}{t_{2}^{1} = 30s} _{53}I^{135} \xrightarrow{\beta, \gamma}{t_{2}^{1} = 6.7h} _{54}Xe^{135}$

Te-135 and I-135 together constitute about 6.3% of all fission products. Due to the short half-life of Te-135 we normally consider the whole 6.3% to be produced as I-135. The rate of production of xenon and iodine from fission depends on the fission rate. Thus:

Rate of production of Xe-135 from fission = $\gamma_{Xe} \Sigma_{f} \phi$

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Rate of production $= \gamma_I \Sigma_f \phi$ of I -135 from fission = $\gamma_I \Sigma_f \phi$

where: γ_{Xe} = Fission product yield of Xe γ_{I} = Fission product yield of I Σ_{f} = Microscopic fission cross section ϕ = Average thermal neutron flux

The rate of production of xenon from iodine depends only on the decay of the iodine, thus:

Rate of production of Xe -135 from I -135 = $\lambda_{I}N_{I}$

where: $\lambda_{I} = \text{decay constant for I } -135(S^{-1})$

$$N_{I} = \text{concentration of } I - 135 \left(\frac{a \text{ cons}}{\text{cm}^{3}}\right)$$

a) Radioactive decay as follows:

$$54Xe^{135} \xrightarrow{\beta \gamma} 55Cs^{135}$$

b) Neutron absorption (burnout):

 $_{54}Xe^{135} + _{0}n^{1} \longrightarrow _{54}Xe^{136} + \gamma$

Niether Cs -135 nor Xe -136 are significant neutron absorbers. The removal rates are as follows:

Rate of change of Xe -135 by decay = $\lambda_{Xe}N_{Xe}$

Rate of change of Xe -135 by burnout $q_a Xe_{Xe^{\varphi}}$

where: λ_{xe} = decay constant for Xe -135

N_{Xe} = Concentration of Xe -135

 σ_a^{Xe} = microscopic absorption cross section

 ϕ = thermal neutron flux

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Now we can set up two equations, one which describes the behaviour of xenon and one which describes the behaviour of iodine. The time rate of change of the iodine $(\frac{d}{dt}N_{I})$ is:

$$\frac{d}{dt} N_{I} = \underbrace{\gamma_{I} \Sigma_{f}}{\gamma_{I} \Sigma_{f}} - \underbrace{\lambda_{I} N_{I}}{\gamma_{I} \Sigma_{f}}$$
(1)

Production Loss due from fission to decay

The time rate of changes of the xenon $(\frac{d}{dt} N_{xe})$ is:



We would like to examine the buildup of xenon in the reactor; however, since much of the xenon comes from iodine we must examine the behaviour of iodine first.

Examining equation (1) you can see that if we startup a reactor with no iodine present we will initially have a production term $(\gamma_I \Sigma_f \phi)$ but no loss term since N_I is zero. As iodine is created the loss term grows (N_I is increasing) while production term remains constant (for constant power). Eventually the loss will equal the production and the iodine level will remain constant. Mathematically:

 $\frac{d}{dt} \mathbf{N}_{\mathbf{I}} = \mathbf{0} = \gamma_{\mathbf{I}} \Sigma_{\mathbf{f}} \mathbf{0} - \lambda_{\mathbf{I}} \mathbf{N}_{\mathbf{I}}$

 $\lambda_{\mathbf{I}} \mathbf{N}_{\mathbf{I}} = \gamma_{\mathbf{I}} \Sigma_{\mathbf{f}} \phi$

$$\underbrace{\sum_{i=1}^{N_{I}} = \frac{\gamma_{I} \Sigma_{f} \phi}{\lambda_{I}}}_{Equili-}$$

brium iodine concentration. This is a simple exponential buildup which can be considered to reach equilibrium after about five half-lives or 30 hours (within 3%). The buildup is shown in Figure 1.



Figure 1

Now we are able to examine the behaviour of xenon. The buildup of xenon is somewhat more complex than the build-up of iodine. Again at equilibrium, $\frac{d}{dt} N_{Xe} = 0$, and production equals loss. Also iodine must be in equilibrium so $\gamma_I \Sigma_f \phi = \lambda_I N_I$. Thus we can write:

$$\gamma_{Xe} \Sigma_{f} \phi + \gamma_{I} \Sigma_{f} \phi = \lambda_{Xe} N_{Xe} + q_{a} N_{Xe} \phi$$
(3)

Rearranging:

 $\underbrace{N_{Xe}}_{\text{Equili-}} = \frac{(\gamma_{Xe} + \gamma_{I}) \Sigma_{f} \phi}{\lambda_{Xe} + \sigma_{a}^{Xe} \phi}$ Equili- $\lambda_{Xe} + \sigma_{a}^{Xe} \phi$ brium Xe concentration

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The buildup is shown graphically in Figure 2.

Figure 2

It again takes about 5 half-lives to reach equilibrium and for xenon this is about 50 hours.

It is useful to know the relative importance of the production and loss terms for xenon at equilibrium. Examining equation (3) we see the relative importance of the production terms depend only on their respective fission product yields. Thus direct production of Xe-135 from fission is about 5% of the total production at equilibrium while indirect production from the decay of I-135 is 95% of the total production.

In examining the loss terms, note that the loss due to decay depends only on the decay constant (λ_{Xe}) . The loss due to burnout depends on the cross section (σ_a^{Xe}) and the neutron flux. Therefore, the relative importance of the loss terms varies from reactor to reactor depending on the normal flux levels. For a given reactor the relative importance varies with power level. For our larger reactors (Bruce and Pickering) full power flux is $7 \times 10^{13} \frac{n-cm}{cm^3 s}$

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Thus; $\gamma_{Xe} = 2.1 \times 10^{-5} s^{-1}$

$$\sigma_a^{Xe} = 24.5 \times 10^{-5} \text{ s}^{-1}$$

Therefore, burnout constitutes more than 90% of the loss at full power.

Reactivity

The reactivity worth of xenon (called Xenon Load) is a function of the concentration of xenon. As it is the reactivity due to xenon that we are concerned about, it is normal to express Xenon Load in reactivity units (Δk_{Xe}) . As shown in Figure 3, the equilibrium Xenon Load for 100% is about -28 mk.

It is also common practice to express the concentration of iodine as Iodine Load in mk. It is important to realize that iodine is not itself a poison hence there is no actual reactivity associated with it. *Iodine Load* is by definition the reactivity if all the iodine present were instantaneously changed to xenon. I repeat it is not an actual reactivity.

By examining the equations for equilibrium xenon and iodine it can be deduced that equilibrium Iodine Load is a direct function of power (eg, doubling the power doubles the Iodine Load) whereas equilibrium Xenon Load does not have such a straightforward relationship with power. Figure 3 shows the approximate variation of equilibrium Xenon Load with power for Bruce or Pickering. The significant point is that equilibrium Xenon Load doesn't change much over the normal operating range.



Figure 3

Xenon Simulation

The reactivity due to Equilibrium Xenon is easily compensated for by designing the reactor to have sufficient excess positive reactivity to overcome the negative reactivity due to the xenon. Now the regulating system must be capable of controlling the excess positive reactivity when there is no xenon present (eg, startup after a long shutdown). This is most commonly done by dissolving a poison (boron or gadolinium) in the moderator and removing it as the xenon builds up. This addition of poison to the moderator on startup is called Xenon Simulation.

As you may suspect, the buildup to and presence of Equilibrium Xenon does not present a significant problem in the operation of our reactors. However, the transient behaviour of xenon creates a major obstacle to operation.

Transient Xenon Behaviour

Assume a reactor has been operating at 100% power long enough for xenon to have reached equilibrium. If power is rapidly reduced to essentially 0%, what happens to the xenon concentration? To answer this question we shall examine the differential equation which describes the time behaviour of xenon.

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$$\frac{d}{dt}N_{xe} = \gamma_{xe}\Sigma_{f}\phi + \gamma_{I}N_{I} - \gamma_{xe}N_{xe} - \sigma_{a} \qquad Xe_{N}_{xe}\phi$$
5% 95% 10% 90%

The percentages shown are the relative magnitudes of the production and loss terms prior to the decrease in power. When power is reduced to 0%, the small production term $(\gamma_{Xe} \Sigma_{f} \phi - direct fission production)$ and the large loss term $(\sigma_{a}^{Xe} N_{Xe} \phi - burnup)$ both cease. Since the major production

term $(\lambda_I N_I - \text{decay of iodine})$ continues the concentration of xenon starts to increase. The increase can't go on forever since there is a limited supply of iodine, thus the xenon peaks and eventually decays away. This is shown graphically in Figure 4.



Figure 5

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Time to Poison Versus Size of Step Reduction in Power from an Initial Power Level of 200 MWe. Values for Other Initial Power Levels are Shown in Tabular Form. Fuel Assumed to be at Equilibrium Irradiation.

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40 MAXIMUM LOAD TO AVOID POISONING OWABLE OUT AL 38 INITIAL POWER ŝ (35[°] (WILLI-K) REACTIVIT MAXIMUM XENON LOAD APPROX. EXCESS W. EQUILIBRIUM LOAD FOR 200 MWe 26 25 24 23 22 21 SIZE OF STEP REDUCTION IN POWER (MW) 20 Q 10 20 30 40 50 60 70 80 90

Maximum Xe load attained during the transient following step reductions in power from various initial power levels. (Xenon Assumed to be at the Corresponding Steady State Value Initially; Fuel Assumed to be at Equilibrium Irradiation).

Figure 7

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removal by neutron capture will also decrease by 40% from its full power value, but because xenon is still being removed the transient will not reach its shutdown peak. Looking at the figure, you will see that for a 40% reduction the available excess reactivity of ~10 mk is just sufficient to override the transient altogether. Ultimately equilibrium will be restored and the xenon load will then be that corresponding to 60% of the full power flux. The figure also shows that the rate of xenon build-up is less for a 60% reduction than for a 100% reduction, and that the poison override time would therefore be longer.

- Fig. 6 shows that this is true, namely that for a fixed amount of excess reactivity the poison override time depends on the size of the power reduction. For example, the curve shows that for a reduction of 120 MWe this time will be 1 hour, but it will be twice that for a reduction of 100 MWe.
- Fig. 7 shows the maximum xenon loads reached during the transient following step reductions from various initial power levels. For example, if the reactor is operating at 160 MWe and is then taken down to 100 MWe, the xenon load will increase from 27.2 mk to 35.1 mk. With 10 mk excess reactivity there should be no problem, but without looking at curves like this you wouldn't know whether there would be.

The converse to these curves also applies. For example, if the reactor is running at 140 MWe (at equilibrium) and it is taken to 200 MWe, the immediate effect will be a gain in reactivity due to increased burnup of xenon. At the same time more iodine will be produced which will not show up as extra xenon production until later on. As a result, the curve will run through a minimum, and than the xenon production will increase because of the increasing amount of iodine that is decaying. Eventually, the xenon concentration will attain the new equilibrium value corresponding to operation at 200 MW. The whole process is shown schematically in Fig. 8, and it does not normally present any operational problems.

Xenon Oscillations

So far, we have assumed that the xenon poisoning and reactivity loads apply to the reactor as a whole. No mention has been made of the possibility of localized changes in xenon poisoning which can have a very important effect on reactor stability.



Xenon Transient Following Step Load Increase From Various Initial Powers to 100% Final Power.

Figure 8

For example, let us consider a reactor that has been operated at power long enough for the iodine and xenon concentrations in the fuel to have reached equilibrium.

Suppose now that without changing the total power of the reactor, the flux is increased in one region of the reactor and simultaneously decreased in another region. This change from the desired normal distribution is called a *flux tilt*. This may happen, for example, if control rods or similar mechanisms are inserted into one region and at the same time withdrawn from another. In the region of increased flux, the xenon now burns out more rapidly than it did prior to the change, and its concentration decreases. This decrease in xenon concentration leads to a higher reactivity in this region, which, in turn, leads to an increased flux. This again leads to increased local xenon burnup, increased local reactivity, increased flux, and so on.

Meanwhile, in the region of decreased flux, the xenon concentration increases due to its reduced burnup and to the continued decay of the existing iodine which was produced in the original, higher flux. This increased xenon concentration decreases the reactivity in this region, which reduces the flux, in turn, increasing the xenon concentration, and so on. The thermal flux, and hence the power density, thus decreases in this region while it increases in the other, the total power of the reactor remaining constant. These local power excursions do not continue forever. In the region of increased flux, the production of xenon from the decay of iodine, which is now being formed more rapidly in this region, ultimately reduces the reactivity there and the flux and power eventually decrease. Likewise, in the region of reduced flux, the accumulated xenon eventually decays, increasing the local reactivity and reversing the flux and power transient in that region.

In this way, the flux and power of a reactor may oscillate between different regions (end to end or side to side) unless action is taken to control them. Calculations, fortunately too lengthy to be spewed out here, show that these *xenon oscillations* have a period of from about 15 to 30 hours.

Xenon oscillations can only occur in large reactors. The argument to show this is as follows:

If the neutrons produced in one region of the reactor do not cause significant fissions in another region, then the two regions can act independently of one another. The criterion that determines whether or not this is possible is the degree of neutron leakage from the one region to the other. In a reactor such as NPD the core is small enough to permit a disturbance started in one region to have an effect in another region. The xenon and flux changes would therefore affect the whole core and a regulating system based on flux measuremen in one locality can correct the flux disturbance and prevent xenon oscillations from being initiated.

If the reactor is large, leakage of neutrons between regions is very small. A disturbance started in one region has little effect in another region. Thus, if a flux increase occurs due to a fuel change in one region, for example, a nonregional regulating system would compensate for this and maintain steady power by lowering the flux in another region to keep the average flux across the core constant. This would set up a xenon oscillation in the second region exactly out of phase with that in the first region.

Furthermore, it is obvious that xenon oscillations can only occur if the flux is high enough for xenon burnup to be as pronounced as xenon decay.

These two conditions for the presence of xenon oscillations (ie, large reactor size and high flux) are satisfied for most power reactors. Since xenon oscillations can occur at constant power they may go unnoticed unless the flux and/or power density distributions are monitored at several points in the reactor. This must be done in order to prevent such oscillations, since they represent something of a hazard to the safe operation of a reactor. Conceivably, they may lead to dangerously high local temperatures and even to fuel meltdown.

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In any event, these oscillations, if permitted to continue, burden the core materials with unnecessary temperature cycling which may result in premature materials failure.

One of the purposes of the regional absorber rods at Douglas Point, and of the regional liquid zone control systems at Pickering and Bruce, is to prevent such xenon oscillations. For example, at Pickering the reactors are subdivided into 14 regions (called zones), and each region has flux detectors whose output is used to adjust the amount of light water absorber in the zone control compartments.

Samarium-149

Sm-149 is the most important of the stable fission products. It is formed in the fuel by the decay of fission product neodymium-149 and promethium-149:

$$Nd^{1+9} \xrightarrow{\beta^{-}} Pm^{1+9} \xrightarrow{\beta^{-}} Sm^{1+9}$$

Since Sm-149 is stable, the only removal process for it is neutron capture. The Sm-150 formed has a low absorption and is therefore insignificant. Sm-149 has a much lower cross section $(4.2 \times 10^{4} \text{b})$ than Xe-135, it will take correspondingly longer for equilibrium to be reached. The half-life of neodymium is so short compared to promethium that we lump its fission product yield with promethium. Note that there is no direct production of samarium from fission. As with xenon we need two equations to describe the behaviour:

$\frac{d}{dt}$ N _{Pm}	$= \gamma_{\mathbf{Pm}} \Sigma_{\mathbf{f}} \phi -$	$\overset{\lambda_{\mathbf{Pm}}N_{\mathbf{Pm}}}{\underbrace{}}$
	Production from fission	Loss due to decav

$\frac{d}{dt}$ ^N Sm	$= \underbrace{{}^{\lambda} Pm^{N}Pm}_{}$	$\sigma a Sm_N Sm^{\phi}$
	Production from decay fo Pm	Loss due to burnup

where:
$$\gamma_{Pm}$$
 = Fission Product Yield of Promethium
 Σ_{f} = Fission Cross Section of the Fuel
 ϕ = Average Neutron Flux
 λ_{Pm} = Decay Constant of Promethium
 N_{Pm} = Number Density of Promethium
 N_{Sm} = Number Density of Samarium
 σ_{a}^{Sm} = Cross Section of Samarium

The equilibrium Pm-149 concentration is:

$$N_{Pm} = \frac{\gamma_{Pm} \Sigma_{f} \phi}{\lambda_{Pm}}$$

Just as with iodine-135, the equilibrium concentration of Pm-149 is a direction function of the power level.

The equilibrium concentration of Sm-149 is:

$$N_{Sm} = \frac{\gamma_{Pm} \Sigma_{f}}{\sigma_{Sm}}$$

Note that equilibrium samarium is independent of the flux level. Equilibrium Samarium Load is around -5.5 mk and it takes about 300 hours of operation to reach equilibrium for our reactors (time to reach equilibrium is a function of the flux level).

Samarium Growth After Shutdown

After a shutdown the samarium concentration will increase since none is being burned out and some is still being produced by the decay of Promethium. The maximum Samarium Load after shutdown depends on the promethium concentration prior to shutdown. For our larger reactors the maximum Samarium Load is about 12 mk. The buildup is shown in Figure 9.



It is interesting to note that although the equilibrium samarium load has to be allowed for in reactor design, the shutdown load may be ignored. There are two reasons for this.

- (1) By looking at the time scale of Fig. 9 you will realize that the maximum samarium load will not appear until the xenon transient has long been and gone. There will therefore be lots of reactivity available. You can also see that the increase in samarium load during the xenon poison override time is negligible, so that this doesn't present a problem either.
- (2) The rate at which the samarium is formed is governed by the Pm-149 half-life of 53 hours, and it corresponds almost exactly to the rate at which Pu-239 is formed after a shutdown. (Pu-239 is still produced from Np-239 decay, but it is not being used up since there are no neutrons). It turns out that the increased reactivity from this plutonium transient more than compensates for the increased samarium load. The net change after shutdown is about +6 mk due to combined effect of samarium and plutonium.

ASSIGNMENT

- 1. Write the equations for the time rate of change of N_{Xe} and N_{I} . Explain what each term represents and give the magnitudes of the terms. Note the conditions under which these magnitudes are applicable.
- 2. Explain why equilibrium Xenon Load changes very little when power is raised from 50% to 100%.
- Explain why peak xenon after shutdown from 100% equilibrium will be nearly twice what it is after shutdown from 50% equilibrium.
- 4. Give and explain the conditions required for a xenon oscillation to occur.
- 5. Define Iodine load and explain its significance.
- 6. Explain why samarium growth after shutdown may be neglected in reactor design.

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