Analysis and Methods of Solving the Bateman Equations for Xe-135 Concentration to Model Neutron Poisons in Various Fission Reactor Systems

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1. Fission Reactor Mechanics and Neutron Poisons

As of April 2024, there are currently over 400 operating fission reactors, which produce nearly 10% of the world's electricity with a combined capacity of 390 MWe. Most of these reactors are thermal reactors, requiring a moderator to slow neutrons to acceptable speeds to take advantage of a greater nuclear cross-section of fission. For example, the cross-section of fission for thermal neutrons can be up to 10^3 times greater than the cross-section of fission for fast neutrons. As a result, thermal reactors can make better use of fissile $^{235}_{92}$U since it more readily undergoes fission with neutrons close to 0.025 eV.

In the fission process, mean total energy release is approximately 200 MeV per reaction, of which 190 MeV is usable.

Table 1: List of fission products of $^{235}_{92}$U and their mean energy

type	MeV
Kinetic energy of fission products	166
Kinetic energy of neutrons	5
Energy of gamma rays emitted at reaction	7
Energy of gamma rays emitted by unstable fission products	7
Kinetic energy of beta particles	5
Kinetic energy of neutrinos	10

From *Nuclear Energy* (5th ed., p. 71), R Murray, 2000, Butterworth-Heinemann.

As opposed to natural decay chains, the products of the fission reaction, which carry most of the energy, cannot so easily be described by a simple branching ratio. The distribution of fission products is bimodal, lending to the fact that the asymmetric reaction tends to favor products with Z approximately equal to some nuclear "magic numbers," which contain complete shells and have greater binding energy per nucleon than their neighbors.

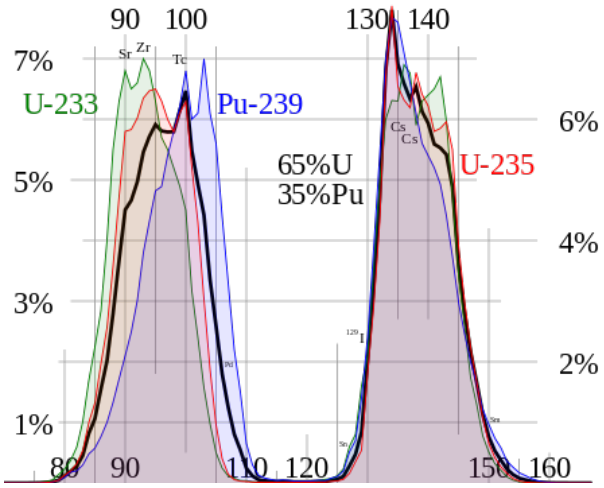

Figure 1: Fission product yield for various fissile nuclei

Additionally, the half-lives of these decay products vary from very short-lived products, such as $^{135}_{52}$Te, with a half-life of 19 seconds, to long-lived products such as $^{93}_{40}$Zr, with a half-life of 1.53 million years.

These decay products tend to saturate in the fuel rods, increasing in concentration until reaching secular equilibrium, when the production rate is roughly equal to that of the total decay rate. While most of these fission products have little effect on the behavior of the remaining neutrons, some have a high absorption cross-section. Additionally, multiple nuclei can undergo processes (such as neutron capture or beta decay) which transmute them into nuclei with larger cross-sections.

Table 2: Sample of fission products and their thermal neutron absorption cross-sections

Nuclide	Cross-Section (barns)	Yield in $^{235}_{92}$U fission
55-Cesium-133 → 55-Cesium-134[1]	140	6.7896 %
53-Iodine-135 → 54-Xenon-135[2]	2650000	6.3333 %
43-Technetium-99	22.8	6.1385 %
40-Zirconium-93	0.696	6.2956 %
61-Promethium-147	168.4	2.2713 %
38-Strontium-90	0.0104	5.7518 %
62-Samarium-149	40720	1.0888 %
53-Iodine-129	30.3	0.9 %

[1] Stable $^{133}_{55}$Cs has a neutron capture cross-section of 29 barns, converting it to $^{134}_{55}$Cs
[2] $^{135}_{55}$I decays with a short half-life to $^{135}_{54}$Xe, also with a relatively short half-life

Of particular note are $^{135}_{54}$Xe and $^{149}_{62}$Sm, known as "neutron poisons", which have massive cross-sections of absorption. In comparison, the cross-section for $^{235}_{92}$U at 0.0253 eV is approximately 500 barns. As $^{135}_{54}$Xe and $^{149}_{62}$Sm accumulate in a reactor to a secular equilibrium, the rate at which they affect the reactivity of the fuel increases as well.

This paper seeks to discuss the effect of these neutron poisons (principally $^{135}_{54}$Xe) on safe reactor operation and implement various methods of computing the concentration of $^{135}_{54}$Xe as a function of time depending on certain reactor characteristics.

2. Modeling Nuclide Concentration

The Bateman equations, first proposed by physicist Ernest Rutherford and solved analytically by mathematician Harry Bateman in the early 20th century, can be used to model the activity of radioactive nuclei of a specific decay series as a function of time. This set of linear, first-order ODEs can be applied to model the concentrations of nuclides in the $^{135}_{54}$Xe decay series described below.

$$^{135}_{52}\mathrm{Te} \xrightarrow{\beta^-} {}^{135}_{53}\mathrm{I} \xrightarrow{\beta^-} {}^{135}_{54}\mathrm{Xe} \xrightarrow{\beta^-} {}^{135}_{55}\mathrm{Cs}$$

For this decay series, the concentration of $^{135}_{54}$Xe and $^{135}_{53}$I can be modeled as:

$$\frac{\mathrm{d}}{\mathrm{d}t}\mathrm{I}(t) = \lambda_{\mathrm{T}}\mathrm{T}(t) - \lambda_{\mathrm{I}}\mathrm{I}(t) \tag{1}$$

$$\frac{\mathrm{d}}{\mathrm{d}t}\mathrm{Xe}(t) = \lambda_{\mathrm{I}}\mathrm{I}(t) - \lambda_{\mathrm{Xe}}\mathrm{Xe}(t) \tag{2}$$

where:

λ_{T} is the decay constant for $^{135}_{52}$Te,
λ_{I} is the decay constant for $^{135}_{53}$I,
λ_{Xe} is the decay constant for $^{135}_{54}$Xe,
$\mathrm{T}(t)$ is the number of $^{135}_{52}$Te nuclei at time t
$\mathrm{I}(t)$ is the number of $^{135}_{53}$I nuclei at time t
$\mathrm{Xe}(t)$ is the number of $^{135}_{54}$Xe nuclei at time t

While equations (1) and (2) hold if there is no other production or consumption of the nuclides under consideration, these equations should be rewritten to involve the production by fission for both $^{135}_{53}$I and $^{135}_{54}$Xe, and consumption via neutron absorption for $^{135}_{54}$Xe. Because $^{135}_{52}$Te has a half-life of just 19 seconds, it makes more sense to consider $^{135}_{53}$I as the primary fission product, and include the fission yield of $^{135}_{52}$Te with that of $^{135}_{53}$I (as is done in Table 2), and remove the $\lambda_{\mathrm{T}}\mathrm{T}(t)$ term when rewriting equation (1).

2.1 Physical Application of the Bateman Equations

The equations for the derivatives of $\mathrm{I}(t)$ and $\mathrm{Xe}(t)$ are therefore:

$$\frac{\mathrm{d}}{\mathrm{d}t}\mathrm{I}(t) = \underbrace{\gamma_{\mathrm{I}}\Sigma_{\mathrm{f}}\varphi(t)}_{\text{fission yield}} - \underbrace{\lambda_{\mathrm{I}}\mathrm{I}(t)}_{\beta^-\text{decay of } {}^{135}_{53}\mathrm{I}} \tag{3}$$

$$\frac{\mathrm{d}}{\mathrm{d}t}\mathrm{Xe}(t) = \underbrace{\gamma_{\mathrm{Xe}}\Sigma_{\mathrm{f}}\varphi(t)}_{\text{fission yield}} + \underbrace{\lambda_{\mathrm{I}}\mathrm{I}(t)}_{\beta^-\text{decay of } {}^{135}_{53}\mathrm{I}} - \underbrace{\lambda_{\mathrm{Xe}}\mathrm{Xe}(t)}_{\beta^-\text{decay of } {}^{135}_{54}\mathrm{Xe}} - \underbrace{\mathrm{Xe}(t)\sigma_{\mathrm{a}}^{\mathrm{Xe}}\varphi(t)}_{\text{neutron absorption}} \tag{4}$$

with the following physical constants defined as:

Table 3: Constants and for the Bateman Equations

Symbol	Definition	Value
λ_{I}	decay constant for ${}^{135}_{53}\mathrm{I}$	$2.9306 * 10^{-5}\ \frac{1}{\mathrm{s}}$
λ_{Xe}	decay constant for ${}^{135}_{54}\mathrm{Xe}$	$2.1066 * 10^{-5}\ \frac{1}{\mathrm{s}}$
γ_{I}	combined fission yield for ${}^{135}_{52}\mathrm{Te}$ and ${}^{135}_{53}\mathrm{I}$	0.0629
γ_{Xe}	fission yield for ${}^{135}_{54}\mathrm{Xe}$	0.002576
σ_{f}	microscopic cross section of fission in ${}^{235}_{92}\mathrm{U}$	$585.1 * 10^{-24}\ \mathrm{cm}^2$
$\sigma_{\mathrm{a}}^{\mathrm{Xe135}}$	microscopic cross-section of absorption in ${}^{135}_{54}\mathrm{Xe}$	$2.65 * 10^{-18}\ \mathrm{cm}^2$
E_f	average energy released per ${}^{235}_{92}\mathrm{U}$ fission	$3.20 * 10^{-11}\ \mathrm{J}$
$N_{\mathrm{U235}}V$	number of ${}^{235}_{92}\mathrm{U}$ atoms in core	$5.13 * 10^{27}$
N_{U235}	density of ${}^{235}_{92}\mathrm{U}$ atoms	$4.74 * 10^{20}\ \frac{1}{\mathrm{cm}^3}$[1]
ν	average number of neutrons released per fission	2.43
Σ_f	macroscopic cross-section of fission in ${}^{235}_{92}\mathrm{U}$	$\sigma_f * N_{\mathrm{U235}}\ [\frac{1}{\mathrm{cm}}]$
ν	average number of neutrons released per fission	2.43

[1] Computed for a sample reactor with 100 metric tons of Uranium fuel at 2% enrichment

Table 4: Variables for the Bateman Equations

Symbol	Definition	Value
$\varphi(t)$	neutron flux as a function of time	$\frac{P}{E_f\sigma_f N_{U235}V}$
P	power generation of the nuclear reactor	0, 1500, or 3000 $\mathrm{MW_T}$
$p_{Xe}(t)$	poison reactivity as a function of time	$-\frac{\sigma_{\mathrm{a}}^{\mathrm{Xe135}}\mathrm{Xe}(t)}{\nu\Sigma_f}$

The formula for poison reactivity is effectively the fraction of fission neutrons absorbed by ${}^{135}_{54}\mathrm{Xe}$. At reactor start-up, concentrations of ${}^{135}_{54}\mathrm{Xe}$ and ${}^{135}_{53}\mathrm{I}$ are both 0. As the power (and accordingly the neutron flux) is increased to the maximum value (3000 $\mathrm{MW_T}$), both concentrations approach a constant value (secular equilibrium). This time can be referred

to as t_s, and the values of the concentrations can be determined given that their rate of change should both be zero. The equilibrium values for both nuclides are:

$$\mathrm{I}(t_s) = \frac{\gamma_\mathrm{I}\Sigma_\mathrm{f}\varphi(t_s)}{\lambda_\mathrm{I}} \tag{5}$$

$$\mathrm{Xe}(t_s) = \frac{(\gamma_\mathrm{I} + \gamma_\mathrm{Xe})\Sigma_\mathrm{f}\varphi(t_s)}{\lambda_\mathrm{Xe} + \sigma_\mathrm{a}^{\mathrm{Xe135}}\varphi(t_s)} \tag{6}$$

Note that the neutron flux may be considered as time dependent, as adjusting the neutron flux can be done in order to reduce power at a given time.

Equations (3) and (4) can be solved with initial conditions $\mathrm{I}(0) = 0$ and $\mathrm{Xe}(0) = 0$ resulting in the following graph, demonstrating the secular equilibrium being reached within approximately 48 hours of operation.

$$\mathrm{I}(t) = \mathrm{I}(t_s)(1 - e^{-\lambda_\mathrm{I} t}) \tag{7}$$

$$\mathrm{Xe}(t) = \frac{\gamma_\mathrm{I}\Sigma_\mathrm{f}\varphi(t_s)}{\lambda_\mathrm{Xe} + \sigma_\mathrm{a}^{\mathrm{Xe135}}\varphi(t_s)}(1 - e^{-\lambda_\mathrm{I} t}) + \frac{\gamma_\mathrm{Xe}\Sigma_\mathrm{f}\varphi(t_s)}{\lambda_\mathrm{Xe} + \sigma_\mathrm{a}^{\mathrm{Xe135}}\varphi(t_s)}(1 - e^{-\lambda_\mathrm{Xe} + \sigma_\mathrm{a}^{\mathrm{Xe135}}\varphi(t_s)t}) \tag{8}$$

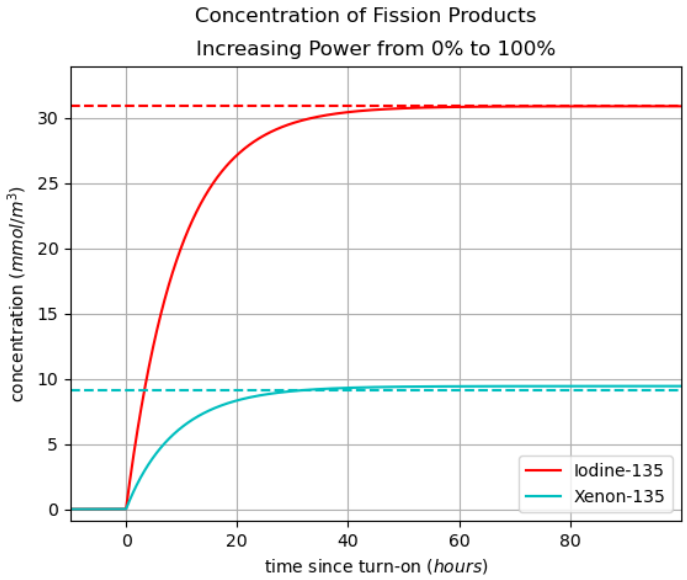

Figure 2: Concentration of fission products after reactor start (3000 $\mathrm{MW_T}$)

Upon reaching secular equilibrium, the concentrations of the nuclides change little. At this point, the effect of neutron poisoning can be readily accounted for, since the reactor is in a stable state. However, if the power to the reactor is dramatically reduced (which can be modeled by setting neutron flux to 0), the $^{135}_{54}\mathrm{Xe}$ concentration spikes, because its production rate exceeds the consumption rate for a brief period of time. The equations for this behavior are below, where equations (3) and (4) have been solved with the initial condition equal to the equilibrium condition.

$$\mathrm{I}(t) = \mathrm{I}(t_s)e^{-\lambda_\mathrm{I} t} \tag{9}$$

$$\mathrm{Xe}(t) = \mathrm{Xe}(t_s)e^{-\lambda_{\mathrm{Xe}}t} + \frac{\mathrm{I}(t_s)\lambda_{\mathrm{I}}}{\lambda_{\mathrm{I}} - \lambda_{\mathrm{Xe}}}(e^{-\lambda_{\mathrm{Xe}}t} - e^{-\lambda_{\mathrm{I}}t}) \tag{10}$$

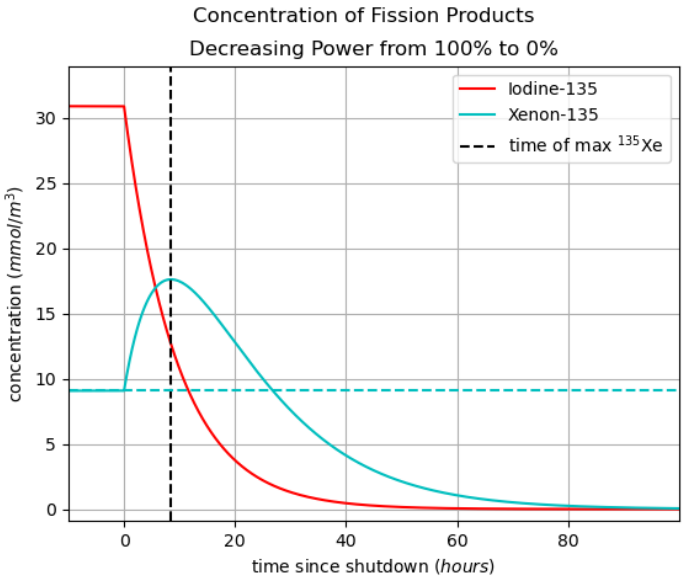

Figure 3: Concentration of fission products after reactor shutdown (3000 $\mathrm{MW_T}$)

For equations (9) and (10), and in figure 3 above, t denotes the time after reactor shutdown. The time at which maximum $^{135}_{54}\mathrm{Xe}$ concentration occurs can be found by computing when $\frac{\mathrm{d}}{\mathrm{d}t}\mathrm{Xe}(t) = 0$; in this case, at roughly 8.45 hours following shutdown.

Both concentrations converge to 0 as time moves on. If power is reduced to approximately 50%, instead of being shut down, the neutron flux decreases to this percentage of its initial maximum as well. However, the fission product concentration will not be halved, due to the decrease in absorption with a smaller neutron flux.

3. Using Numerical Methods to Model Flux Change

To simplify calculations, an explicit solution to equations (3) and (4) need not be derived. Implementing a numerical method such as the forward Euler Method, with an adjusted value for the neutron flux, while not as fast computing the analytic solution, is still not particularly computationally expensive. It is also worth noting that the Forward Euler Method is stable only for sufficiently small Δt. The Forward Euler Method is described below for timestep Δt, and is implemented for equations (3) and (4).

$$y(t_{n+1}) = y(t_n) + \Delta t \frac{\mathrm{d}y}{\mathrm{d}t}(t_n) \tag{11}$$

$$\mathrm{I}(t_{n+1}) = \mathrm{I}(t_n) + \Delta t(\gamma_{\mathrm{I}}\Sigma_{\mathrm{f}}\varphi(t_n) - \lambda_{\mathrm{I}}\mathrm{I}(t_n)) \tag{12}$$

$$\mathrm{Xe}(t_{n+1}) = \mathrm{Xe}(t_n) + \Delta t(\gamma_{\mathrm{Xe}}\Sigma_{\mathrm{f}}\varphi(t_n) + \lambda_{\mathrm{I}}\mathrm{I}(t_n) - \mathrm{Xe}(t_n)(\lambda_{\mathrm{Xe}} + \sigma_{\mathrm{a}}^{\mathrm{Xe}}\varphi(t_n))) \tag{13}$$

To represent a decrease in power from 3000 to 1500 $\mathrm{MW_T}$, $\varphi(t_n)$ can be fixed to $\frac{\varphi(t_s)}{2}$. The following figure is generated from using the Forward Euler method to plot a reduction in power of a 3000 $\mathrm{MW_T}$ reactor from 100% to 50%.

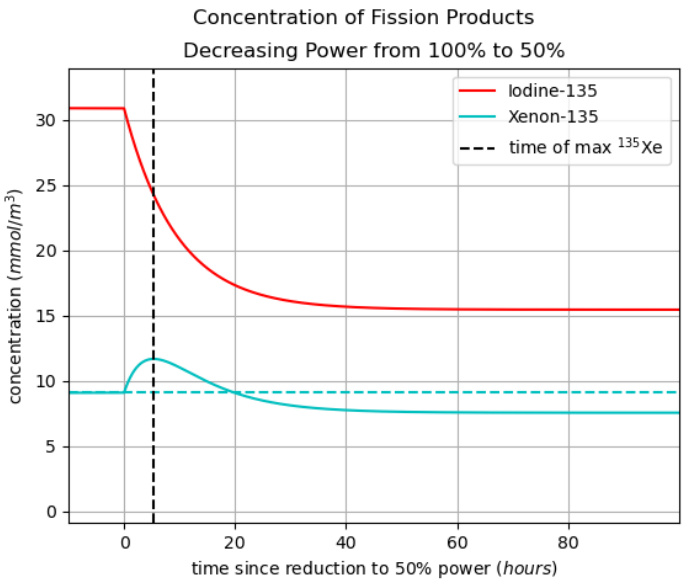

Figure 4: Concentration of fission products after decreasing power to 1500 $\mathrm{MW_T}$

The same method can be implemented to model the removal of control rods at 50% power, with the intent of bringing the power (and neutron flux) to its previous maximum of 100%. Equations (11) and (12) are implemented again using the Forward Euler method, with $\varphi(t_n) = \varphi(t_s)$.

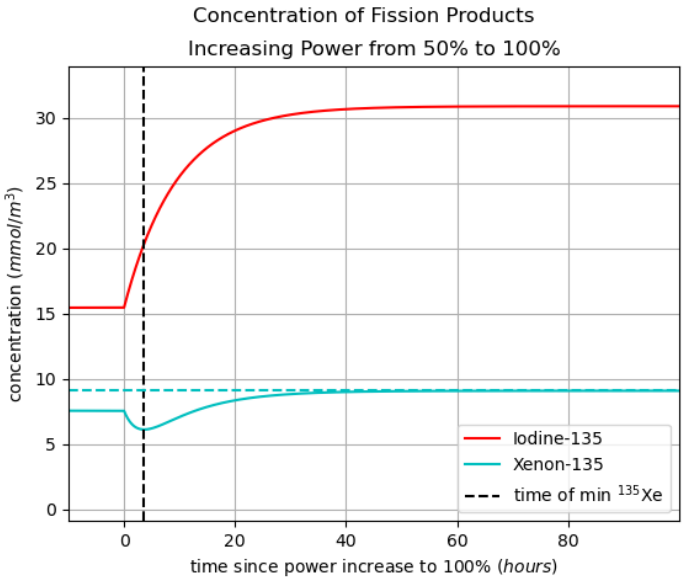

Figure 5: Concentration of fission products after increasing power to 3000 $\mathrm{MW_T}$

Both figures 4 and 5 demonstrate essential aspects of nuclear poisons, by which following a decrease in the power of the reactor, $^{135}_{54}\mathrm{Xe}$ concentration rises. In effect, this is a positive feedback loop by which reactivity is lessened until the concentration of xenon decreases.

The poison reactivity as a function of time can be computed from the formula in Table 4. The figures below demonstrate the change in reactivity corresponding to the relative concentration of $^{135}_{54}$Xe at specific time values.

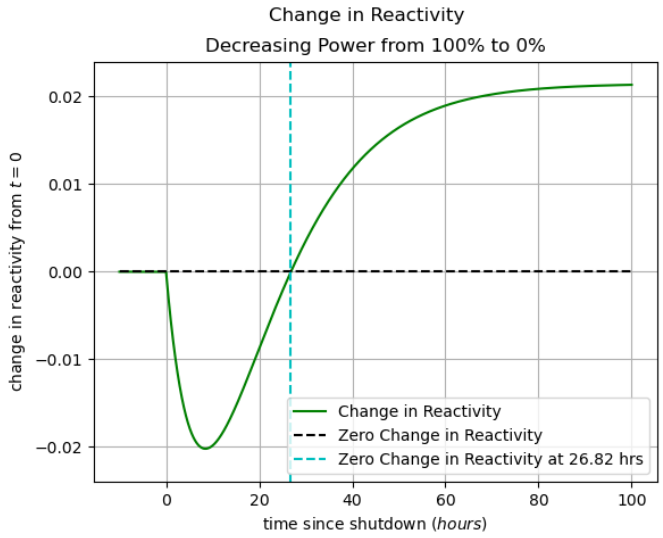

Figure 6: Poison Reactivity of $^{135}_{54}$Xe when shutting reactor off

In this case, the "xenon pit" can be observed, by which the reactivity drops for a period of time after shutting the reactor off. In Figure 6, the time for the poisoning effect to be reduced back to a baseline level is over 26 hours.

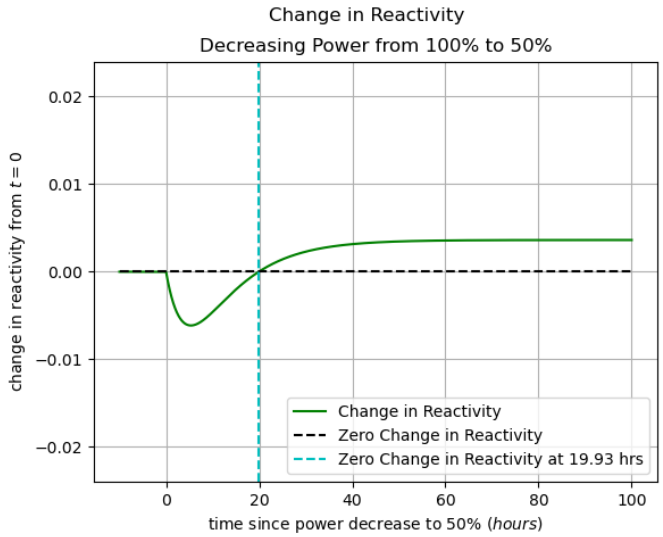

Figure 7: Poison Reactivity of $^{135}_{54}$Xe when decreasing reactor power from 100% to 50%

The same effect can be observed for a power decrease to only 1500 MW_T, by which the effect of Xenon is relevant for up to 19.93 hours after the change in neutron flux.

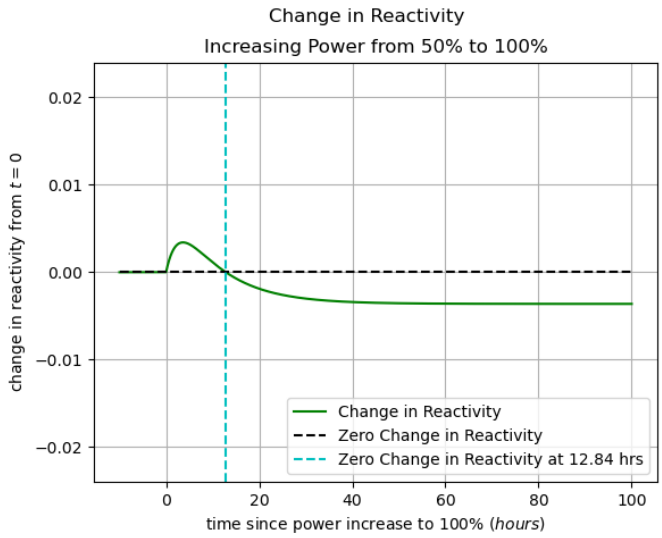

Figure 8: Poison Reactivity of $^{135}_{54}$Xe when increasing reactor power from 50% to 100%

This figure demonstrates the great safety risk of improperly accounting for $^{135}_{54}$Xe. When the power is increased from 1500 MW_T to 3000 MW_T, the concentration of $^{135}_{54}$Xe decreases, effectively reducing the amount of neutrons taken up by $^{135}_{54}$Xe, and enabling a greater fission rate.

4. Analysis and Discussion

In the case described in Figure 8, if this behavior is not accounted for by operators, reactivity can spike above a critical value. Among other engineering and structural failures, this effect contributed to the criticality incident causing an explosion at the Chernobyl Power Plant in the USSR, 1986.

In practice, the Bateman equations provide a useful (but quite simplified) tool to model nuclear poisons. In further studies, cross-sections could be adjusted for other fuel compositions and reactor designs. Additionally, other nuclear poisons such as $^{149}_{62}$Sm could be considered for a more comprehensive treatment of safe reactor operation.

Lastly, although the Forward Euler method converges for the small timestep used in this study, other numerical methods could be implemented for solving this problem if other adjustments to the Bateman equations are made.

Note: programs and resources available at the public github repository here

5. References

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