Physical Mechanisms for Fission Product Concentration Differences from Variations in Reactor Power History

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Abstract

A method for uniquely determining power history characteristics of spent fuel assemblies based on the concentration of various fission products in the fuel assembly has been developed. It is envisioned that this method could be used as a transparency aid at reprocessing facilities to verify the identity of spent fuel assemblies. Specific fission product concentrations measurably vary as a result of differences in reactor power history. A discussion of the physical properties of various fission products responsible for concentration differences along with the mechanisms by which these properties produce concentration differences is presented. Several different mechanisms based on variations in the physical properties are illustrated through simplified models. These models led to the identification of an extensive list of possible monitor ratios which is presented. A case study was also performed to assess the distinguishing capabilities of the given monitors. The variations between cases include modifications to specific power, the number of shutdowns, and the duration of the shutdowns while maintaining a constant final burnup. For all but one of the cases a monitor ratio is shown to vary by at least 20% with the base case while some ratio differences for cases with simple modifications reached 300%. This paper concludes with a proposed technique using the presented monitor ratios to verify the identity of spent fuel assemblies based on differences in reactor power history.

Introduction

The power history of a spent fuel assembly may be used to uniquely identify the assembly. The power history experienced by a fuel assembly is dependent on core power, assembly position within the core, and other factors. These dependencies result in a unique power history for each assembly. A method to verify the power history of a fuel assembly could provide additional transparency for international safeguards applications. Further, if such a method were based on fission product signatures, the method's susceptibility to deception would be minimized. The use of fission products to determine spent fuel parameters such as actinide concentrations, fuel burnup, fuel age, reactor type, fuel type, and initial fuel enrichment has been demonstrated previously. [1-8] There has also been some preliminary work on power history determination from fission products.[9] Building from the prior fission product analyses, research was conducted to determine a method for power history identification based on fission product signatures.

Analytical Models

In order to determine ideal monitor properties analytical models of potential monitor isotopes were constructed. Both radioactive and stable monitors were considered for this analysis.

A model was derived for radioactive fission product monitors following the assumed decay scheme shown in Figure 1(a). The radioactive monitor is produced directly from fission with an arbitrary decay constant, λ_R, and neutron absorption cross-section, $\sigma_{a,R}$. The concentration of the radioactive monitor, N_R, for any time t within a given time step is given by:

$$N_R^i(t) = \frac{\rho_U P_s^i}{\overline{E_R}\lambda_{R,eff}}\left[\frac{Y_{f,U235}^R}{1+R^{-1}} + \frac{Y_{f,Pu239}^R}{1+R}\right]\left[1-e^{-\lambda_{R,eff}(t-t_{i-1})}\right] + N_R^{i-1}e^{-\lambda_{R,eff}(t-t_{i-1})}, \text{ for } t_{i-1} < t < t_i \quad (1)$$

where $\lambda_{R,eff} = \lambda_R + \frac{\sigma_{a,R}\rho_U P_s^i}{\overline{E_R}\overline{\Sigma_f}}$, $R \equiv \frac{\sigma_{f,U235}\overline{N_{U235}}}{\sigma_{f,Pu239}\overline{N_{Pu239}}}$, ρ_U is the density of uranium in the fuel, P_s^i is the specific power of timestep i, $\overline{E_R}$ is the average energy released per fission, $\overline{\Sigma_f} = \sigma_{f,U235}\overline{N_{U235}} + \sigma_{f,Pu239}\overline{N_{Pu239}}$ is the average macroscopic fission cross-section, $\sigma_{f,X}$ is the microscopic fission cross-section of isotope x, $Y_{f,X}^R$ is the fission yield of the radioactive fission product isotope from the fission of isotope x, $\sigma_{a,R}$ is the neutron absorption cross-section of the radioactive fission product isotope, and λ_R is the decay constant of the radioactive isotope. Equation 1 is piecewise continuous over i timesteps for which specific power is assumed constant.

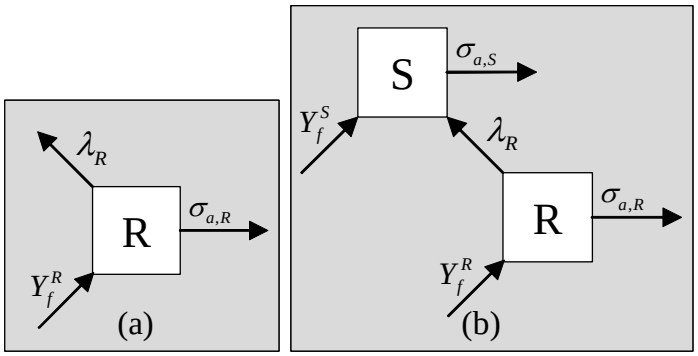

Figure 1. Assumed Decay Schemes for (a) Radioactive Fission Product Monitor and (b) Stable Fission Product Monitor.

A model was derived for a stable fission product monitor in which the stable monitor is produced directly from fission and through the decay of a single radioactive parent isotope. The radioactive parent isotope is also produced directly from fission and has a decay constant, λ_R, and neutron absorption cross-section, $\sigma_{a,R}$. The stable monitor isotope also has a neutron absorption cross-section, $\sigma_{a,S}$. This decay scheme is shown in Figure 1(b). The concentration of the stable monitor, N_S, is then given by

$$N_S^i(t) = \frac{\rho_U P_s^i}{\overline{E_R}\sigma_{a,S,eff}}\left[\frac{Y_{f,U235}^S}{1+R^{-1}} + \frac{Y_{f,Pu239}^S}{1+R}\right](1-e^{-\sigma_{a,S,eff}(t-t_{i-1})}) +$$

$$\lambda_R\left\{\begin{array}{l}\frac{\rho_U P_s^i}{\overline{E_R}\lambda_{R,eff}}\left[\frac{Y_{f,U235}^R}{1+R^{-1}} + \frac{Y_{f,Pu239}^R}{1+R}\right]\left[\frac{(1-e^{-\sigma_{a,S,eff}(t-t_{i-1})})}{\sigma_{a,S,eff}} + \frac{(e^{-\sigma_{a,S,eff}(t-t_{i-1})} - e^{-\lambda_{R,eff}(t-t_{i-1})})}{\left(\sigma_{a,S,eff} - \lambda_{R,eff}\right)}\right] \\ + \frac{N_R^{i-1}(t_{i-1})(e^{-\lambda_{R,eff}(t-t_{i-1})} - e^{-\sigma_{a,S,eff}(t-t_{i-1})})}{\left(\sigma_{a,S,eff} - \lambda_{R,eff}\right)}\end{array}\right\} \qquad (2)$$

$$+N_S^{i-1}(t_{i-1})e^{-\sigma_{a,S,eff}(t-t_{i-1})}, \text{ for } t_{i-1} < t < t_i$$

where $\sigma_{a,S,eff} = \frac{\sigma_{a,S}\rho_U P_s^i}{\overline{E_R}\overline{\Sigma_f}}$.

These models were tested using simple fuel burnup scenarios to ensure that the models replicated expected results.

Reactor Power History Variation Scenarios

The models above were used to understand how the decay constant and cross-sections influence the concentration of potential monitors in response to power history variations. Table 1 shows the ranges of values used for the decay constants and cross-sections of the potential monitor models in this analysis.

Table 1. Ranges of Parameters Varied for Potential Monitor Models

	Radioactive Monitor	Stable Monitor
Parent Half-Life [yr]	N/A	0.0055-40,000
Parent Cross-section [b]	N/A	1-10,000
Monitor Half-Life [yr]	0.0055-40,000	N/A
Monitor Cross-section [b]	1-10,000	1-10,000

While varying the parameters as described in Table 1, both models were applied to power history scenarios which varied specific power and shutdown time. The total burnup for the power history scenarios was held constant at 35 GWd/MTU. The first power history scenario consisted of a single irradiation cycle with a constant specific power and no shutdowns. The constant specific power was varied from 1–110 W/g. The second power history scenario consisted of three irradiation cycles of equal burnup with variable specific power and no shutdowns. The specific powers of the first and third cycles were held constant at 100 W/g while the second cycle specific power was varied from 10–100 W/g. The third power history scenario involved three burn cycles of equal burnup divided by shutdowns of variable duration. The total shutdown time was varied between 20 d – 2000 d and evenly divided between the two shutdowns. The specific power of each cycle for the third scenario was held constant at 35 W/g.

Results

Differences in final monitor concentrations generated through the power history variation analyses led to identification of ideal values of monitor properties. Useful ranges of monitor properties are shown in Figure 2.

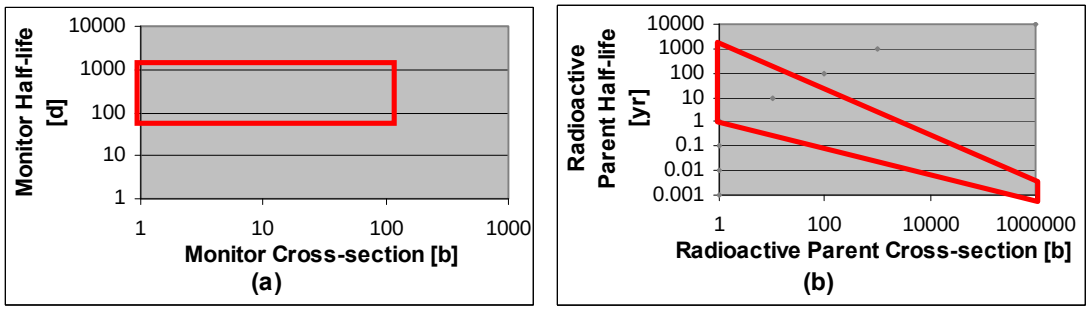

Figure 2. Useful property ranges for (a) potential radioactive monitors and (b) potential stable monitors.

Discussion

The ranges of useful monitor properties is determined by the value of $\lambda_{R,eff}$, $\sigma_{a,S,eff}$, and the time duration of the irradiation. Since in all scenarios the final burnup was constant, the total amount of monitor isotope atoms produced was also constant. However, the amount of each isotope lost via absorption or decay or produced via parent decay varies depending on the specific power history. The implications of $\lambda_{R,eff}$ will be explored separately for the radioactive monitor and the stable monitor.

Radioactive Monitor

Concentration differences in radioactive monitors from variations in power history were found to be a result of an optimal destruction mechanism of the monitor. The destruction mechanism of the radioactive monitor is controlled by the effective decay constant $\lambda_{R,eff}$. If $\lambda_{R,eff}$ is too small, a negligible amount of destruction will occur and concentration will not vary with power history. However, if $\lambda_{R,eff}$ is too large the monitor will decay too rapidly leading to saturation of the monitor at the current specific power and loss of measurable signal after shutdown. A Figure of Merit (*FOM,R*) was defined for the radioactive monitor as

$$FOM,R = \frac{N_R^i\left(t_i, P_s^i = 10\text{ W/g}\right) - N_R^i\left(t_i, P_s^i = 100\text{ W/g}\right)}{N_R^i\left(t_i, P_s^i = 10\text{ W/g}\right)} * 100 * N_R^i\left(t_i, P_s^i = 10\text{ W/g}\right) \quad (3)$$

where the power history consists of a single irradiation cycle to 35 GWd/MTU. This *FOM,R* accounts for the increased percent difference for large values of $\lambda_{R,eff}$. This *FOM,R* is plotted against $\lambda_{R,eff}$ in Figure 3.

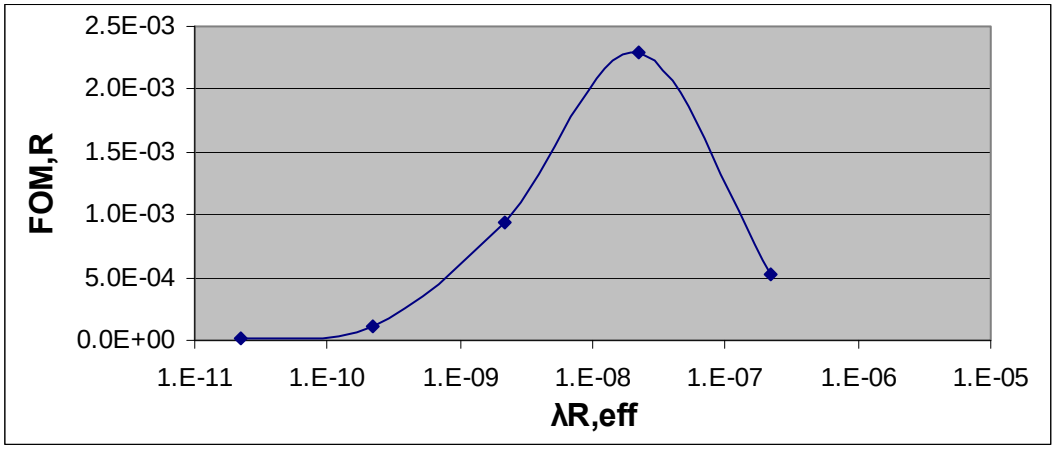

Figure 3. Optimal range of $\lambda_{R,eff}$ for a radioactive monitor

The limits shown in Figure 2(a) can be derived in part from Figure 3. The limits of $\lambda_{R,eff}$ may be directly applied to the monitor half-life. The influence of cross-section on radioactive monitor concentration is not as clear as cross-section coupled to specific power as seen in the definition of $\lambda_{R,eff}$. Substituting for specific power by $P_s^i = BU_i\left(t - t_{i-1}\right)^{-1}$ in Equation 1 and referring to the product $\lambda_{R,eff}\left(t - t_{i-1}\right)$ one sees that if decay is negligible relative absorption, irradiation time will cancel:

$$\lambda_{R,eff}\left(t - t_{i-1}\right) = \frac{\sigma_{a,R}\rho_U BU_i}{\overline{E_R}\,\overline{\Sigma_f}}.$$

The result is that variations in specific power will not produce variations in monitor concentration. If decay is not relatively negligible then a large cross-section will shift $\lambda_{R,eff}$ above the useful range. As such minimal cross-sections are desirable and correspond to the limits of cross-section shown in Figure 2(a). It is noteworthy that the value of this optimal $\lambda_{R,eff}$ is approximately inversely proportional to the reactor period. It was seen that if reactor period is increased the limits of $\lambda_{R,eff}$ will be decreased.

Stable Monitor

Concentration differences in stable monitors from variations in power history were found to be a result of an optimal production mechanism from the decay of the monitor's radioactive parent. Production via parent decay is represented by the second term in Equation 2. In order for concentration differences to occur in the stable monitor, the rate of production via decay of the parent must be sufficiently limited so that irradiation time (as specified by specific power) determines the amount of production via decay. If the rate of decay is too fast, total time will not impact the amount of production. The properties of the radioactive parent determine the amount of stable monitor production via decay.

For an appropriate discussion of how the properties of the radioactive parent limit production via decay the parent destruction rate $\lambda_{R,eff}$ must be resolved into its components of decay, λ_R, and absorption, $\sigma_{a,R}$. λ_R directly limits production via decay. There is thus a range of λ_R for which production will be sufficiently limited but not so limited that production will be negligible. The percent difference between final monitor concentrations produced from a single irradiation cycles to 35 GWd/MTU with specific powers of 10 W/g and 100 W/g is plotted against λ_R in Figure 4 for two values of $\sigma_{a,R}$: (a) $\sigma_{a,R}$ = 0.001 b and (b) 10,000 b.

Neutron absorption by the radioactive parent serves as competition for production via decay. This competition serves to drive the useful range of λ_R to higher values. This is illustrated in Figure 4(b) by increasing the value of $\sigma_{a,R}$ to 10,000 b. The optimal range of parent half-life and $\sigma_{a,R}$ shown in Figure 2(b) reflect the useful range of λ_R as influenced by $\sigma_{a,R}$. Another affect of the competition provided by absorption is a boost in concentration experienced by the stable monitor as a result of reactor shutdowns.[6] This concept is illustrated by Figure 5.

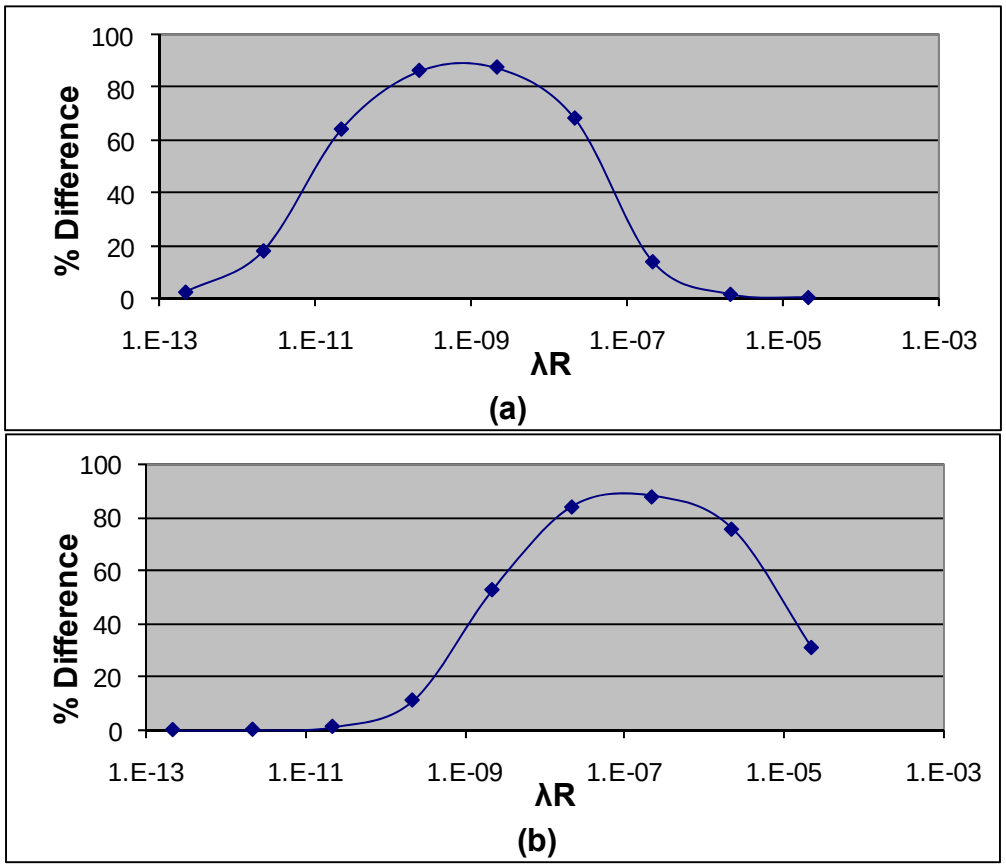

Figure 4. Optimal range of λ_R for a stable monitor with (a) $\sigma_{a,R}$ = 0.001 b and (b) 10,000 b

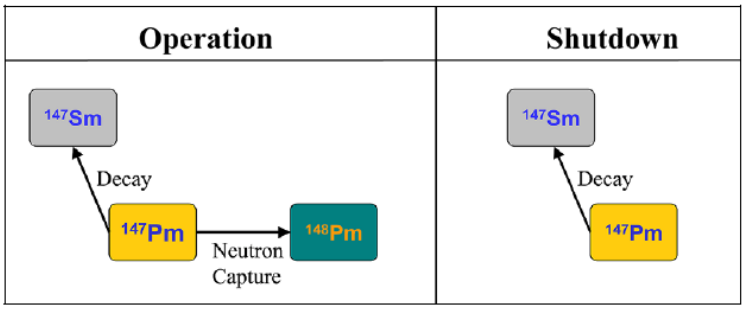

Figure 5. Illustration of significant parent absorption cross-section leading to an increase of stable monitor concentration as a result of reactor shutdown. Figure taken from ref. 6

If cross-section is large enough to move the useful parent property range to the far right of Figure 2(b), the presence of reactor shutdowns will no longer be reflected in the stable monitor concentration. In this case the useful parent half-life is so short that all parent atoms will immediately decay if not absorbed. Such a monitor would reflect only variations in non-zero specific power. The parent daughter pair of Xe-135 and Cs-135 exhibit the properties required of a specific power only monitor. Comparison of the Cs-135 concentration and the Sm-147 concentration (depicted in Figure 5) may directly isolate total shutdown time.

Conclusion

Isotopic concentration differences will result from variations in specific power when the destruction mechanism, for radioactive isotopes, or the production mechanism via parent decay, for stable isotopes, is optimized such that the concentration of the isotopes is influenced by the corresponding variations in time. The research presented has identified a range of values of the isotopic properties necessary to optimize the destruction and production mechanisms to generate significant monitor isotope concentration differences in response to power history variations.

Using the range of ideal monitor properties, specific monitor ratios were identified for use as power history characterization signatures. Identified radioactive monitor ratios include Te-127m/Te-125, Ce-144/Ce-140, and Sn-123/Sn-126. Identified stable monitor ratios include Eu-151/Eu-153, Gd-155/Gd-157, Gd-154/Gd-157, Sm-147/Sm-150, and Cs-135/Cs-133. Further, double ratios of a stable monitor ratio and a ratio of the monitors radioactive parent were found to significantly increase the signature differences between power history cases. These signatures are recommended based on numerical differences of monitor ratios produced by a case study of several reactor power histories simulated with TransLat. [9,10]. The results showed variations of the recommended signatures between the base case and all but one of the other power history cases of 20% – 300%.

Implementation of a system using the recommended power history signatures to verify the identity of spent fuel assemblies could enhance international nuclear safeguards. Such a system would begin with a sample of a spent fuel assembly dissolution. Initially all other obtainable fuel parameters such as burnup, fuel age, and initial enrichment should be determined from the fuel sample. With knowledge of those fuel parameters, physics parameters such as averaged fission yields and averaged cross-sections can be obtained from physics codes such as TransLat. Using the power history declared by the reactor operator, the fuel parameters, and the models in Equation 1

and Equation 2, estimates for monitor ratios may be made. These estimates would then be verified against the measured monitor ratios for confirmation of spent fuel assembly identity.

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