

A MODEL FOR PREDICTING FISSION PRODUCT ACTIVITIES IN REACTOR COOLANT: APPLICATION OF MODEL FOR ESTIMATING I-129 LEVELS IN RADIOACTIVE WASTE

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ABSTRACT

A general model was developed to estimate the activities of fission products in reactor coolant and hence to predict a value for the I-129/Cs-137 scaling factor; the latter can be applied along with measured Cs-137 activities to estimate I-129 levels in reactor waste.

The model accounts for fission product release from both defective fuel rods and uranium contamination present on in-core reactor surfaces. For simplicity, only the key release mechanisms were modeled. A mass balance, considering the two fuel source terms and a loss term due to coolant cleanup was solved to estimate fission product activity in the primary heat transport system coolant. Steady state assumptions were made to solve for the activity of short-lived fission products. Solutions for long-lived fission products are time-dependent.

Data for short-lived radioiodines I-131, I-132, I-133, I-134 and I-135 were analysed to estimate model parameters for I-129. The estimated parameter values were then used to determine I-129 coolant activities. Because of the chemical affinity between iodine and cesium, estimates of Cs-137 coolant concentrations were also based on parameter values similar to those for the radioiodines; this assumption was tested by comparing measured and predicted Cs-137 coolant concentrations.

Application of the derived model to Douglas Point and Darlington Nuclear Generating Station plant data yielded estimates for I-129/I-131 and I-129/Cs-137 which are consistent with values reported for pressurised water reactors (PWRs) and boiling water reactors (BWRs). The estimated magnitude for the I-129/Cs-137 ratio was 10^{-8} - 10^{-7} .

INTRODUCTION AND BACKGROUND

Scaling factors are generally employed to estimate the concentrations of DTM radionuclides. They relate the activity of DTM radionuclides to the activity of easy-to-measure (ETM) gamma-emitting marker radionuclides such as Co-60 and Cs-137. Scaling factors for several radionuclides of interest to OPG, namely, Pu-238, Pu-239, Pu-240, Pu-241, Am-241, Cm-242, Cm-244, Fe-55, Ni-63, C-14 and Sr-90 have been under development (1) since 1999. However, the development of experimental scaling factors for I-129, a potential key radionuclide, poses a major challenge because of its very low concentration in reactor waste. Theoretical modeling approaches are, therefore, of interest.

Modeling approaches for predicting the concentrations of fission-based radionuclides (e.g. I-129, Tc-99) have been applied in the literature. For instance, the French have developed the PROFIP code (2) for the estimation of fission product and actinide concentrations in the primary

system of pressurised water reactors (PWRs). Similarly, Tractebel, Belgium (3) has developed a predictive capability for several radionuclides based on various inputs including the concentrations of Co-60, Cs-137 and short-lived radioiodines. Unlike the European codes, Vance and Associates in the US have developed a code called 3R-STAT (4) (it superseded an earlier code called Radsorce (5)) which focussed on the predictions of only I-129 and Tc-99 levels in the coolant. Similar to the European codes, however, 3R-STAT predictions are based on the measured concentrations of Co-60, Cs-137 and short-lived radioiodines in the coolant.

While the European codes are proprietary, details of 3R-STAT are readily available. A review of 3R-STAT indicated the modeling treatment to be exceedingly complex. The complexity results from the inclusion of several mechanisms for radionuclide releases from both defective and tramp fuel; some of the mechanisms are relatively unimportant and could be eliminated from the formulation without a significant impact. It was felt that a new model based only on a consideration of the key release processes could be developed into a predictive capability not only for I-129 and Tc-99 but also for other fission products of interest. A model development addressing the estimation of I-129 and Cs-137 concentrations in the coolant and hence the I-129/Cs-137 scaling factor, is presented in this paper. Note that the scaling factor estimated from coolant concentrations of I-129 and Cs-137 is assumed to be directly applicable to radioactive waste; departure from this assumption can possibly be accounted for empirically.

This paper presents an overview of the model and provides a comparison of model predictions with literature data. Unless literature references are specifically cited in this paper, the reader must assume that the specifics are drawn from a detailed paper being published elsewhere (6).

OVERALL APPROACH FOR PREDICTING I-129 LEVELS IN WASTE

Fission products are released into the primary heat transport (PHT) system coolant both from defective fuel rods and from uranium contamination present on in-core reactor surfaces. The release mechanisms are briefly discussed in Section 3.1. As a result of the fuel-based releases, significant levels of various fission products such as iodines, xenons, kryptons and cesiums are observed in reactor coolant. OPG stations routinely monitor the levels of short-lived radioiodines, namely, I-131, I-132, I-133, I-134 and I-135 in the PHT coolant to detect the presence of failed fuel in reactor channels (in addition, levels of several other radionuclides including Cs-137 and Co-60 are also recorded).

A model based on a description of the steady state release mechanisms for short-lived radioiodines was used to apportion the observed radioiodine releases into its two source term contributions (defected and tramp fuel) in order to assess the characteristics of failed fuel. The characteristics of the failed fuel were deduced from the magnitudes of model parameters obtained by fitting the observed radioiodine data to the model. Note that the relative contribution of defected and tramp fuel would depend on the number of defective fuel rods and the extent of tramp fuel residing in-core.

Because isotopic differences are considered to be generally negligible, estimates for model parameters, obtained by fitting the short-lived radioiodine data are also applicable for estimating I-129 releases. Unlike in the case of short-lived radioiodines, however, the radioactive decay term in the mass balance for long-lived species is negligible and hence steady state is not achieved (for a given radionuclide, steady state occurs, as a rule of thumb, after an operating period of 3 to 3.5 half-lives); the release of long-lived I-129 from defective fuel, therefore, requires the solution of time-dependent diffusion equations.

While the predicted concentration of I-129 in the coolant can be used to estimate the integrated releases from fuel (released I-129 is subsequently captured by the ion exchange resins in the PHT purification circuit), it cannot be directly used to estimate the inventory of I-129 in individual waste packages. For this purpose, it is desirable to predict the concentration of I-129 relative to that of Cs-137 in the coolant, i.e., the value of the scaling factor I-129/Cs-137. Similar to I-129, Cs-137 is also relatively long-lived and hence its release from defected fuel is also time dependent. Note that it is preferable to estimate the I-129/Cs-137 ratio from the model rather than combine the theoretical concentration estimate for I-129 with the measured concentration of Cs-137 in the coolant. The latter approach leads to difficulties because plant Cs-137 data is influenced by fueling/defueling operations and possibly other factors.

While the time-dependent diffusion equation is applicable for modeling the releases of both I-129 and Cs-137 from defected fuel, model parameter values for the two nuclides may differ. In the absence of an independent means for estimating the model parameters for Cs-137, their values as a first approximation were assumed to be identical or similar in magnitude to those established for I-129 from short-lived radioiodines data. This assumption is reasonable based on the observation that cesium is chemically associated with iodine in the fuel-sheath gap. The assumption was further tested by comparing the magnitude of the predicted and measured Cs-137 concentrations in the coolant.

In addition to radioiodine releases during normal reactor operation, significant and potentially larger radioiodine releases may occur during reactor shutdown as a result of iodine spiking. This occurs because iodine deposits, which form on internal fuel and clad surfaces during reactor operation, may be mobilized from defected fuel by condensing water following a shutdown. Releases during shutdown were also modeled; because the releases occur rapidly (within hours), the process was modeled by assuming it to be instantaneous.

SUMMARY OF MODEL DEVELOPMENT

Relative Importance of Release Mechanisms

Fission products are released in to the PHT system coolant from two sources, namely, 1) from defects in fuel elements and 2) from tramp uranium. The mechanisms for release are:

- ◆ Solid state lattice diffusion - this mechanism is temperature dependent
- ◆ Direct recoil - release from external surfaces can occur by direct recoil when a fission fragment, with sufficient kinetic energy is produced within a surface layer less than or equal to its range in the solid (the range for the ~ 67 MeV iodine fission fragment in solid UO₂ is ~8 μm). This mechanism is independent of temperature.
- ◆ Knockout - release from external surfaces can also occur by knockout when either a primary fission fragment or energetic particle created in a collision cascade interacts elastically with a fission product atom. This process is also independent of temperature.

When a fission fragment strikes a stationary atom of the lattice, a primary knock-on may be created. The latter is likely to be a uranium or oxygen atom of the fuel, although occasionally a stationary fission product atom may be dislodged. Similarly, the primary

knock-on can also transfer kinetic energy to other atoms of the lattice by elastic collision, creating secondary and in-turn, higher-order knock-ons. The range of the higher-order uranium knock-on (which has an initial kinetic energy of ~200 eV) in the UO₂ is ~50 Å. To a first approximation, the fission products can be considered to be knocked on in the same manner as the uranium atoms of the lattice.

Releases from defective fuel occur dominantly by diffusion. In operating fuel rods, recoil is not an important release process, except for very short-lived fission products in which case the diffusional contribution may be significantly reduced as a result of decay. In general, recoil release is relatively unimportant because fission fragments, born within a distance of their maximum range from the fuel surface, have sufficient energy to embed themselves in the adjacent fuel cladding; only a very small fraction of the fission fragments (2% for iodine) are actually retained in the fuel to clad gap (typical gap thickness is 20 μm).

In comparison to diffusional release from defective fuel, release via knockout is even less insignificant than releases via recoil.

In contrast, releases from tramp uranium contamination are dominated by the contribution from recoil. Tramp fuel is typically in the form of small spherical particles of diameter ~ 10 μm. As a result of this small particle size, the temperature generated by fission heating is generally too low for diffusion to be an important transport process. In contrast, since any fission fragment generated in the small fuel particle has a sufficiently long range (range of the fission fragment is comparable to the diameter of the fuel particle), it is instantaneously released from the fuel particle by recoil into the coolant.

Releases from tramp fuel via knockout are insignificant because the range of the knockout particle (50 Å) is very small compared to the size of the fuel particle. Further, the release rate for knockout is proportional to the fission product concentration in the fuel particles; unless the fuel debris originates by ejection from a defective fuel element, its fission product concentration is very small and thus the knockout contribution is further reduced.

Release from Defective Fuel into the Fuel-Clad Gap

Considering diffusional transport to be the dominant mechanism, a mass balance on the concentration $C(r,t)$ (atom m⁻³) of a radioactive species for an "idealized" spherical fuel grain of radius a can be represented by the equation

$$\frac{\partial C}{\partial t} = D\nabla^2 C - \lambda C + \beta \quad (\text{Eq. 1})$$

where the fission rate production term β (atom m⁻³ s⁻¹) is offset by losses due to diffusion (D is the diffusion coefficient (m² s⁻¹)) and radioactive decay (λ is the decay constant (s⁻¹)). From Fick's law of diffusion, the release rate from the fuel grain per unit volume, R_f (atom m⁻³ s⁻¹) is given by:

$$R_f = \frac{4\pi a^2}{(4\pi a^3 / 3)} \left(-D \frac{\partial C}{\partial r} \Big|_{r=a} \right) = -\frac{3D}{a} \frac{\partial C}{\partial r} \Big|_{r=a} \quad (\text{Eq. 2})$$

For a short-lived radioactive species, Equation (1) can be simplified by setting the derivative equal to zero. Combining the result with Equation (2) yields the following expression after simplification:

$$\left(\frac{R}{B}\right)_{dif} = 3\sqrt{\frac{D'}{\lambda}} \quad (\text{Eq. 3})$$

where the release rate R (atoms/s) equals $R_f V$ (V is the volume of fuel in m^3), the birth rate B (atoms/s) equals βV and $D' = D/a^2$. The corresponding expression for stable radionuclides (λ equals 0) is given by the expression

$$\left(\frac{R}{B}\right)_{dif} = \left[6\sqrt{\frac{\tau}{\pi}} - 3\tau\right] \quad (\text{Eq. 4})$$

where $\tau = D t$

Release from Fuel-Clad Gap into the Coolant

Volatile fission products released into the fuel-clad gap migrate towards the defect site where they may eventually be released into the reactor coolant. Treating the release as a first-order rate process, the following mass balance equation can be written for the fission product inventory in the gap:

$$\frac{dN_g}{dt} = R_{dif} - (\lambda + \nu)N_g \quad (\text{Eq. 5})$$

where N_g represents the number of atoms in the gap and ν is the gap escape rate coefficient (s^{-1}). Note that the magnitude of ν depends on the size of the fuel defect. Substituting R_{dif} according to Equation (3) and noting that the release rate into the coolant is given by

$$R_c(t) = \nu N_g(t) \quad (\text{Eq. 6})$$

the following expression is obtained for short-lived radioactive species:

$$\frac{R_c}{B} \approx 3\left(\frac{\nu}{\lambda + \nu}\right)\sqrt{\frac{D'}{\lambda}} \quad (\text{Eq. 7})$$

The corresponding expression for long-lived or stable species is given by

$$\frac{R_c(t)}{B} = \left[(1 - e^{-\psi\tau}) - 6\psi \sum_{n=1}^{\infty} \frac{e^{-\psi\tau} - e^{-n^2\pi^2\tau}}{n^2\pi^2(n^2\pi^2 - \psi)} \right] \quad (\text{Eq. 8})$$

where $\psi = \nu D'$ and $\tau = D t$.

Release from Tramp Uranium Fuel into the Coolant

Considering only recoil, the release-to-birth rate for tramp fuel is given by

$$\left(\frac{R}{B}\right)_{rec} = \varepsilon \left(\frac{S_g}{V}\right) \mu_f \quad (\text{Eq. 9})$$

where S_g/V is the geometrical surface area to volume ratio (m^{-1}) for the solid and μ_f is the fission fragment range in solid UO_2 (m) and ε is the release efficiency. Substituting the theoretical expression for the release efficiency, it can be shown that, for surface deposited fuel particles, Equation (9) simplifies to

$$\left(\frac{R}{B}\right)_{rec} = 0.5 \quad (\text{Eq. 10})$$

while for tramp fuel particles in the coolant, the corresponding ratio is unity.

Because fission products are released instantaneously via recoil, Equations (9) and (10) do not have a time dependence as in the case of diffusional transport for stable species.

Combined Release from Defective and Tramp Uranium Fuel for Short-lived Fission Products during Normal Reactor Operation

Assuming steady state, Equation (7) for a short-lived species and Equation (10) can be combined to yield the total release rate into the coolant from defective and tramp fuel:

$$\left(\frac{R}{y}\right)_c = \left(\frac{\nu}{\lambda + \nu}\right) \frac{A}{\sqrt{\lambda}} H + c \quad (\text{Eq. 11})$$

where $A = x(3\sqrt{D'}F_f)$ and $c = \frac{1}{2}F_t$.

Here x is the number of defective fuel rods, F_f is the average fission rate (fission s^{-1}) per defective rod and F_t is the fission rate in the tramp uranium fuel. Note that to obtain Equation (11), B in Equation (7) was replaced by $x F_f y$ while B in Equation (10) was replaced by $F_t y$ where y represents the fission product yield (atoms/fission). The parameter H is a dimensionless correction factor which results from a consideration of the diffusion of a parent-daughter pair¹; an enhanced diffusional release occurs only for radionuclides such as I-132 (where $H \approx 6$) which have a relatively long-lived precursor.

The three unknown parameters ν , A and c in Equation (11) can be obtained by fitting data for at least three short-lived radioiodines after first substituting the total release rate in Equation (11) by the measured rate. The latter is derived from coolant activity measurements based on the following mass balance for the number of atoms of a given species in the coolant (N_c):

$$\frac{dN_c}{dt} = R'_c - (\lambda + \beta_p)N_c \quad (\text{Eq. 12})$$

where R'_c is the total release rate from defective and tramp fuel and β_p is the ion-exchange purification rate constant (s^{-1}) ($\beta_p = f_p \varepsilon_p / M$, where f_p is the cleanup system flow rate (kg/s), ε_p is the cleanup system efficiency and M is the mass of water (kg) in the PHT system). Because the PHT system is a closed system, Equation (12) presumes that other loss terms for the radioactive species, such as through the station stack, are comparatively negligible. Under steady state, the measured release rate is given by:

$$R_{meas} = (\lambda + \beta_p)N_c \Rightarrow \left(\frac{R}{y}\right)_{meas} = \left(\frac{\lambda + \beta_p}{\lambda}\right) \frac{A_c}{y} \quad (\text{Eq. 13})$$

where $A_c (= \lambda N_c)$ is the measured coolant activity (in Bq) for a given radionuclide.

A typical fit on a logarithmic plot using data for the five short-lived radioiodines is shown in Figure 1; this represents a situation where both fuel failures and tramp uranium contamination are present. Note that H , which is approximately equal to 1 for all the short-lived radioiodines except I-132, is implicitly accounted for in the fit by normalising the data point for I-132 (most of the release for I-132 is attributed to diffusional transport from defective fuel of its relatively long-lived precursor Te-132) by a factor of 6. In the region where the R/y curve is independent of the decay constant (in this region, the diffusional transport of I-132, I-134 and I-135 from defective

¹ In addition to production by direct fission, the short-lived radioiodines are also produced by decay of precursor telluriums. The precursors for I-131 ($t_{1/2}=8.04d$), I-132 ($t_{1/2}=2.3 h$), I-133 ($t_{1/2}=20.8 h$), I-134 ($t_{1/2}=52.5 m$) and I-135 ($t_{1/2}=6.6h$) are Te-131 ($t_{1/2}=25 m$), Te-132 ($t_{1/2}=78 h$), Te-133 ($t_{1/2}=12.4 m$), Te-134 ($t_{1/2}=42 m$) and Te-135 ($t_{1/2}=19.2 s$), respectively.

fuel is insignificant because of their relatively short half-lives), the release is derived solely from tramp uranium contamination; this portion of the curve, therefore, yields the value of the parameter c . The slope of the curve at smaller values of λ is typically in the range between -0.5 and -1.5 and indicates the presence of fuel failures (a zero slope in this region would indicate the absence of fuel failures). For a large-sized failure with little holdup in the fuel-to-clad gap, i.e., $\nu \gg \lambda$, and hence a $\lambda^{-1/2}$ dependence results whereas for a tight failure where $\nu \ll \lambda$, a $\lambda^{-3/2}$ dependence is obtained.

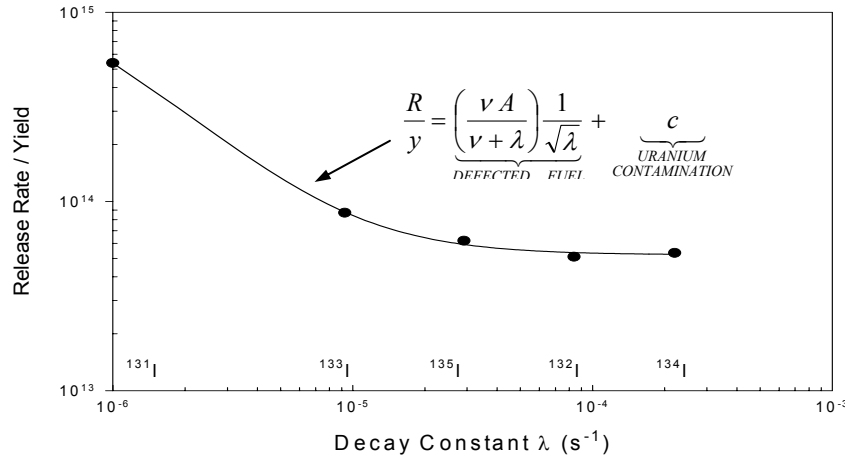


Fig. 1 Plot of Release Rate/Yield versus Decay Constant for Short-Lived Radioiodines - Data for the Douglas Point NGS, March 1982

To estimate D' independently, the average fission rate of the defective rods, F_f , and the number of fuel failures, x , must both be known *a priori*. If such information is unavailable, then D' (s^{-1}) must be estimated from previously established correlations to the average-core linear heat rating P (in kW/m).

Combined Release from Defective and Tramp Uranium Fuel for Long-Lived Fission Products during Normal Reactor Operation

Analogous to Equation (12), the mass balance in the coolant for long-lived species such as I-129 can be represented by

$$\frac{dA_c}{dt} = \lambda R_c^l(t) - \beta_p A_c \tag{Eq. 14}$$

As before, substituting the release rate expressions for defective fuel (a short term ($\tau < 0.1$) approximation for Equations 8 was used) and tramp fuel (Equation 10) into Equation (14) and integrating yields the following:

$$A_c(t) = \mu x F_f y \left\{ \frac{1 - e^{-\phi\tau}}{\phi} + \left(\frac{e^{-\psi\tau} - e^{-\phi\tau}}{(\psi - \phi)} \right) \frac{3}{\psi} \left[1 - \sqrt{\psi} \cot \sqrt{\psi} \right] + 6\psi \sum_{n=1}^{\infty} \left(\frac{e^{-\phi\tau} - e^{-n^2\pi^2\tau}}{n^2\pi^2(n^2\pi^2 - \psi)(n^2\pi^2 - \phi)} \right) \right\} + \lambda c y \left(\frac{1 - e^{-\phi\tau}}{\beta_p} \right) \tag{Eq. 15}$$

where $\mu = \lambda/D'$, $\psi = \nu/D'$, $\phi = \beta_p/D'$, and $\tau = D't$. Equation (15) is based on the short term ($\tau < 0.1$) approximation for R_c^t .

Thus, using the values for the fitted parameters (ν , D' and c) obtained previously from the results for the short-lived iodines, Equation (15) can be used to estimate the activity of long-lived I-129 in the coolant as a function of time.

Combined Release from Defective and Tramp Uranium Fuel for Long-Lived Fission Products After Reactor Shutdown

At shutdown, iodine which had previously deposited in the fuel to clad gap is released essentially instantaneously (via ionic diffusion in liquid water, with possible convective assistance) into the coolant. Thereafter, its concentration in the coolant will decrease as a result of purification flow. Thus, the I-129 concentration in the coolant or equivalently its activity will be described by:

$$N_c(t) = [N_{co} + N_g] e^{-\beta_p t} \Rightarrow A_c(t) = [A_{co} + A_{go}] e^{-\beta_p t} \quad (\text{Eq. 16})$$

where $N_c(t)$ and $A_c(t)$ are, respectively, the I-129 coolant inventory and activity at time t following a shutdown and N_{co} and A_{co} are the corresponding initial quantities at the time of shutdown. These latter quantities can be evaluated from Equation (15). The quantity A_{go} , the initial gap activity at shutdown, follows from Equations (6) and (8) and is given by:

$$A_{go} = \left[\frac{(1 - e^{-\psi\tau})}{\psi} - 6 \sum_{n=1}^{\infty} \frac{e^{-\psi\tau} - e^{-n^2\pi^2\tau}}{n^2\pi^2(n^2\pi^2 - \psi)} \right] \mu x F_f y \quad (\text{Eq. 17})$$

APPLICATION OF MODEL EQUATIONS

Data for Douglas Point Reactor

Available Douglas Point data on the characteristics of failed fuel and the activities of short-lived radioiodines in the coolant were analysed to estimate the activity concentration of I-129 in the coolant according to Equation (15). For the sampling period, ~18 defective fuel rods (at a linear heat rating of about 40 kW/m) operated in the Douglas Point reactor; their average fuel burnup was 110 MWh/kgU. The radioiodine data, earlier shown as Figure 1, are summarised in Table I.

The following parameter estimates were obtained by fitting the above radioiodine data to Equation (11):

$$D' = 6.8 \times 10^{-10} \text{ s}^{-1} \quad (\text{note that } x \text{ is given to be } 18)$$

$$\nu = 1.4 \times 10^{-6} \text{ s}^{-1}$$

$$c = 5.2 \times 10^{13} \text{ fission s}^{-1} \text{ and}$$

$$A = 8.4 \times 10^{11} \text{ fission s}^{-1.5}.$$

Table I Estimated Fission Rates for Short-Lived Radioiodines Based on Measured Activity Concentrations in Douglas Point Coolant

Radioiodine	λ (1/s)	R/y (fission/s)
I-131	9.98×10^{-7}	5.39×10^{14}
I-132	8.37×10^{-5}	5.10×10^{13}
I-133	9.26×10^{-6}	8.72×10^{13}
I-134	2.20×10^{-4}	5.34×10^{13}
I-135	2.91×10^{-5}	6.20×10^{13}

The fuel residence time, t was estimated from the average burnup and linear power, considering that the Douglas Point fuel rod contained 0.710 kgU and had a fuel length of 0.477 m. Thus,

$$t = 110 \text{ MWh/kgU} \times 0.710 \text{ kgU} / (40 \text{ kW/m} \times 0.477 \text{ m} \times 10^{-3} \text{ MW/kW} \times 24 \text{ h/d})$$

$$\sim 170 \text{ d} = 1.47 \times 10^7 \text{ s.}$$

The following additional data were used to estimate I-129 activity according to Equation (15):

$$\lambda = 1.40 \times 10^{-15} \text{ s}^{-1}$$

$$y = 1.08 \times 10^{-2} \text{ atom fission}^{-1} \text{ (this was calculated for a fuel burnup of 110 MWh/kgU)}$$

$$F_f = 1.489 \times 10^{13} \text{ P} = 5.96 \times 10^{14} \text{ fission s}^{-1} \text{ per rod (P = 40 kW m}^{-1}\text{)}$$

$$\beta_p = 8.65 \times 0.99 / 67,000 = 1.28 \times 10^{-4} \text{ s}^{-1} \text{ is assumed in the analysis (where IX purification flow rate is 8.65 L s}^{-1}\text{, column efficiency is 99\%, and PHTS volume is 67,000 L)}$$

Further, the following were estimated:

$$\psi = v/D' = 2059,$$

$$\phi = \beta_p/D' = 1.880 \times 10^5 \text{ and}$$

$$\mu = \lambda/D' = 2.059 \times 10^{-6}.$$

Substituting the values of the above constants and parameters in Equation (15) yielded a predicted I-129 coolant activity of 390 Bq after ~170 d of reactor operation; the estimate is many orders of magnitude smaller than the observed activity for I-131 (1.3×10^{11} Bq). This result, as discussed in Section 4.3, is consistent with the values observed for pressurized water reactors (PWRs) and boiling water reactors (BWRs). The predicted coolant activity of I-129 arose mostly from defective fuel.

The effect of a shutdown on I-129 levels in the coolant was also considered. Based on an initial (after ~170 d of operation) I-129 coolant activity of ~390 Bq ($A_{co} \sim 390$ Bq), Equations (16) and (17) predict a spike resulting in a final coolant activity of $A_{go} \sim 34,770$ Bq. This iodine spike is clearly much greater than the release during normal reactor operation.

Data for Darlington Nuclear Generating Station (DNGS)

Data on average concentrations of various radioiodines in Darlington Nuclear Generating Station (DNGS) coolant (calculated from the 1999 plant data for Units 1-4) were used to estimate the parameters in Equation (11). The estimated values are:

$$D' = 4.0 \times 10^{-10} \text{ s}^{-1}$$

$$\nu = 4.4 \times 10^{-8} \text{ s}^{-1} \text{ and}$$

$$c = 1.8 \times 10^{12} \text{ fission s}^{-1}.$$

The value of D' based on the assumption that only a single fuel defect exists, i.e. $x = 1$ is consistent with the value obtained earlier for Douglas Point data. The fitted value of the gap escape rate constant ($\nu \sim 4.4 \times 10^{-8} \text{ s}^{-1}$), which is significantly lower than that estimated for Douglas Point data, suggests that the failed rod has a tight defect. The fitted parameter $c \sim 1.8 \times 10^{12} \text{ fission s}^{-1}$ for the tramp contribution is a factor of ~ 30 smaller than that estimated for Douglas Point data; this observation is consistent with better fuel management practices in post Douglas Point reactors.

The fuel residence time, t was estimated from the average burnup of 180 MWh/kgU and fuel rod linear power of 39.5 kW/m, considering that the Darlington fuel rod contains 0.511 kgU and has a fuel length of 0.477 m. Thus,

$$t = 180 \text{ MWh/kgU} \times 0.511 \text{ kgU} / (39.5 \text{ kW/m} \times 0.477 \text{ m} \times 10^{-3} \text{ MW/kW} \times 24 \text{ h/d}) \sim 200 \text{ d}.$$

For estimating the activity of I-129, the following additional data were used in Equation (15):

$$\lambda = 1.40 \times 10^{-15} \text{ s}^{-1}$$

$$y = 1.19 \times 10^{-2} \text{ atom fission}^{-1} \text{ (this was calculated for a fuel burnup of 180 MWh/kgU)}$$

$$F_f = 1.489 \times 10^{13} \text{ } P = 5.88 \times 10^{14} \text{ fission s}^{-1} \text{ per rod (} P = 39.5 \text{ kW m}^{-1}\text{)}$$

$$\beta_p = 10 \times 0.99 / 280,000 \sim 4 \times 10^{-5} \text{ s}^{-1} \text{ (where IX purification flow rate is } 10 \text{ kg s}^{-1}\text{, column efficiency is 99\%, and PHTS volume is 280,000 kg)}$$

$$\psi = \nu/D' = 110$$

$$\phi = \beta_p/D' = 1.0 \times 10^5 \text{ and}$$

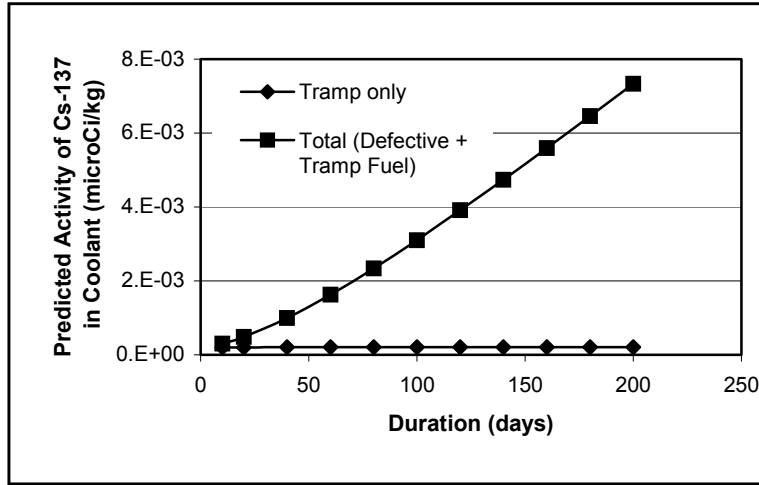
$$\mu = \lambda/D' = 3.5 \times 10^{-6}.$$

Based on the values for the above constants and parameters, the time dependence of the I-129 coolant concentration was predicted using Equation (15). Similarly, the time dependence of the Cs-137 coolant concentration was also estimated for the following two cases:

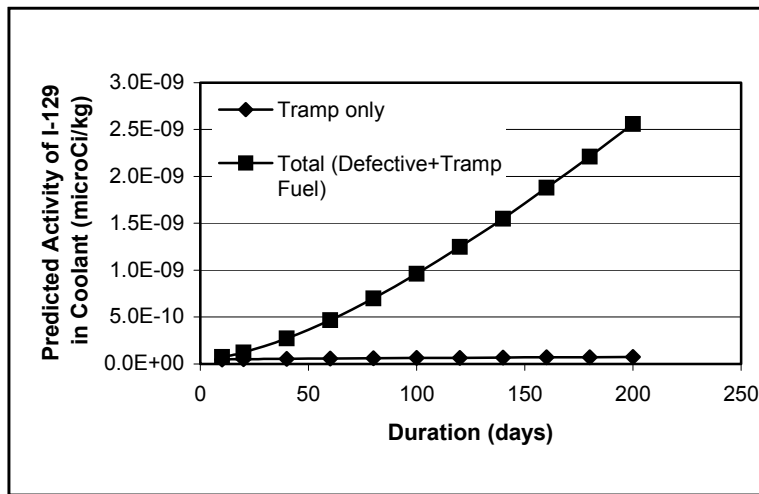
- ◆ Case I : $\nu_{\text{Cesium}} = 3 \nu_{\text{Iodine}}$ and $(\epsilon_p)_{\text{Cesium}} = 0.1 (\epsilon_p)_{\text{Iodine}}$
- ◆ Case II: $\nu_{\text{Cesium}} = \nu_{\text{Iodine}}$ and $(\epsilon_p)_{\text{Cesium}} = (\epsilon_p)_{\text{Iodine}}$

The results for Case II are graphed in Figure 2. Results for both cases indicate the following:

- ◆ Even with only one defected fuel rod, the I-129 releases are dominated by the contribution of defected fuel (except possibly at $t < 10 \text{ d}$).
- ◆ The total I-129 inventory in the coolant at $t = 200 \text{ d}$ equals $280,000 \text{ kg} \times 2.56 \times 10^{-9} \text{ } \mu\text{Ci/kg} \times 3.7 \times 10^4 \text{ Bq/} \mu\text{Ci}$ or only 27 Bq.
- ◆ Similar to I-129, the total Cs-137 release is dominated by the contribution of defected fuel (except possibly at $t < 10 \text{ d}$).



(a)



(b)

Fig. 2. Predicted Activity of a) I-129 and b) Cs-137 as a function of Time for Case II
 $(\nu_{\text{Cesium}} = \nu_{\text{Iodine}} \text{ and } (\epsilon_p)_{\text{Cesium}} = (\epsilon_p)_{\text{Iodine}})$

- ◆ For Case I, the estimated Cs-137 concentration in the coolant of $0.134 \mu\text{Ci/kg}$ or $5.0 \times 10^3 \text{ Bq/kg}$ at $t=200 \text{ d}$ is consistent with the average measured value of $0.15 \mu\text{Ci/kg}$ or $5.6 \times 10^3 \text{ Bq/kg}$. For Case II, however, the estimated Cs-137 concentration in the coolant was a factor of 20 lower than the average measured value.
- ◆ For Case I, estimates of the I-129/Cs-137 ratio, considering only tramp fuel releases are within a factor of 2 of the estimates based on both tramp and defected fuel releases. For Case II, the corresponding estimates are identical in accordance with Equation (18) (see next Section)
- ◆ The I-129/Cs-137 estimates are essentially constant with time but sensitive to the values of the $\nu_{\text{Cesium}}/\nu_{\text{Iodine}}$ and $(\epsilon_p)_{\text{Cesium}}/(\epsilon_p)_{\text{Iodine}}$.
- ◆ The estimated order of magnitude (10^{-8} - 10^{-7}) for the ratio I-129/Cs-137 is consistent with measurements reported in the literature (see next section).

Finally, it was estimated that, as a result of spiking upon shutdown at $t = 200 \text{ d}$, the ratio I-129/Cs-137 for Case I would increase from 1.9×10^{-8} to $\sim 6 \times 10^{-7}$ (a factor of ~ 30). For Case II, the ratio is unaffected by spiking.

Comparison of Model Predictions with Data in the Literature for Normal Operation

Comparisons between measured and predicted I-129/I-131 ratios and between measured and predicted I-129/Cs-137 ratios are shown in Table II. Note the following:

- ◆ The measured EDF data (2) are based on coolant samples from 7 PWR plants.
- ◆ The measured Battelle data (5) are based on 7 resin samples (5 PWRs and 2 BWRs). The NUREG data (7) are based on 32 resin samples, 5 dry active waste samples, 4 filter samples and 1 reactor coolant sample; these samples are drawn from both PWRs and BWRs (note that the Battelle data are included in the NUREG data set). The predicted NUREG data for I-129/Cs-137 ratios are based on radionuclide inventory estimates for used fuel obtained using ORIGEN.
- ◆ The Japanese data are for solidified homogeneous liquid waste (8); the number of samples are not known, and
- ◆ The Kinectrics data (9) is based on an I-129 measurement ($3 \times 10^{-7} \text{ Ci/m}^3$ or $1.1 \times 10^4 \text{ Bq/m}^3$) on one PHT resin sample and a mean Cs-137 concentration ¹¹ of 16 Ci/m^3 or $5.9 \times 10^{11} \text{ Bq/m}^3$.

Table II Comparison Between Measured and Predicted I-129/I-131 and I-129/Cs-137 Ratios

Data Source	Measured*		Predicted	
	I-129/I-131	I-129/Cs-137	I-129/I-131	I-129/Cs-137
EDF, France	2.1×10^{-9}	9.8×10^{-9}	-	-
Battelle, USA	6.5×10^{-9}	1.3×10^{-7}	-	-
NUREG, USA	-	1.2×10^{-7}	-	2.8×10^{-7} (PWRs) 2.9×10^{-7} (BWRs)
JGC, Japan	-	2.5×10^{-8} (PWRs) 5.7×10^{-7} (BWRs)	-	-
Kinectrics	-	1.9×10^{-8}	-	-
This study	-	-	3.0×10^{-9} (Douglas Pt) 2.8×10^{-8} (DNGS)	1.9×10^{-8} (DNGS Case I) 3.5×10^{-7} (DNGS Case II)

*Based on geometric mean values

Despite differences between PWR / BWR and CANDU fuel (the former have considerably longer burnups but much lower heat ratings (~22 kW/m versus ~40 kW/m)), measured I-129/Cs-137 from all sources are mutually consistent and agreement with model predictions is satisfactory. Note in particular the agreement between the Kinectric's measurement data for I-129/Cs-137 and the corresponding predictions for DNGS (Cases I and II). A similar comment applies for the ratio I-129/I-131. The consistency in the model predictions for Douglas Point and Darlington reactors with the reported EDF and Battelle PWR/BWR data provides assurance on the fundamental soundness of the derived model equations.

The NUREG predictions for I-129/Cs-137 are based on ORIGEN code calculations for 500d irradiated used fuel (the estimations varied only slightly with fuel burnup consistent with the calculations for DNGS). The close agreement between measured and estimated scaling factor ratios is indicative of the similar transport behaviours of I-129 and Cs-137 from irradiated fuel. Based on this, values of the various parameters in Equation (15) are likely to be similar for both I-129 and Cs-137 as assumed in the Case II calculations for DNGS.

When parameter values for I-129 and Cs-137 in Equation (15) or Equation (16) are assumed to be identical, the I-129/Cs-137 activity ratio reduces to the simple form:

$$\frac{A_I(t)}{A_{Cs}(t)} = \frac{(\lambda y)_I}{(\lambda y)_{Cs}} \quad (\text{Eq. 18})$$

where the coolant activity ratio is independent of the relative release contributions from defected and tramp fuels and equals the activity ratio in the fuel for both normal and spiking releases. This result explains the agreement between measured and estimated I-129/Cs-137 values observed in the NUREG data set. Note that the weak time or burnup dependence in Equation (18) arises solely from the burnup dependence of the yields for the individual nuclides. It should also be noted that Equation (18) represents the present basis for estimating I-129 inventories in OPG's LLW and ILW.

SUMMARY AND CONCLUSIONS

- ◆ A model was developed to estimate the activities of fission products in reactor coolant. The model accounts for fission product release from both defective fuel rods and uranium contamination that may be present on in-core reactor surfaces. For simplicity, only the key release mechanisms were modeled:
 - Diffusion is considered to be the dominant process for release from the fuel matrix into the fuel-to-clad gap with the subsequent release of fission products from the gap into the primary coolant during normal reactor operation being modeled as a first order rate process; following shutdown, the release of water soluble radionuclides present in the gap inventory is considered to occur instantaneously. This spiking release can cause a several fold increase in the activity burden of specific radionuclides such as I-129.
 - In comparison, only direct recoil is considered to be an important release mechanism for tramp uranium contamination.
- ◆ A mass balance, considering the defective and tramp fuel source terms and a loss term due to coolant cleanup was solved to estimate the activity in the primary heat transport system coolant. Steady state assumptions were made to solve for the activity of short-lived radionuclides. Time-dependent solutions were derived for long-lived radionuclides.
- ◆ The model was applied to estimate the activities of long-lived I-129 and Cs-137 in the coolant and hence to estimate the I-129/Cs-137 scaling factor ratio. For this purpose, data for short-lived radioiodines I-131, I-132, I-133, I-134 and I-135 were analysed to estimate model parameters. The parameter values were then used to estimate I-129 coolant activities. Because of the affinity between iodine and cesium, similar parameter values were assumed to estimate Cs-137 coolant activities; the assumptions were tested by comparing measured and predicted Cs-137 coolant concentrations.
- ◆ Despite differences between PWR / BWR and CANDU fuel, measured I-129/Cs-137 from several data sources are mutually consistent and agreement with model predictions is satisfactory. A similar comment applies for the ratio I-129/I-131. The consistency in the model predictions for Douglas Point and Darlington reactors with measured data provides assurance on the fundamental soundness of the derived model equations. The estimated magnitudes for the I-129/I-131 and I-129/Cs-137 ratios were 10^{-9} - 10^{-8} and 10^{-8} - 10^{-7} , respectively.
- ◆ When identical parameter values are assumed for both the DTM and the ETM radionuclides such as I-129 and Cs-137, respectively, it can be shown that their activity or scaling factor ratio in the coolant is independent of the relative release contributions from defected and tramp fuels and to be equal to the activity ratio in the fuel; this is the case for both normal and spiking releases. This result explains the observed agreement between literature I-129/Cs-137 values for US PWR/BWR waste and those estimated for used fuel using the ORIGEN code. The French I-129/Cs-137 scaling factor data are a factor of ~ 10 lower than the data for US plants and suggest that the assumptions of identical parameter values for both I-129 and Cs-137 may not always apply. Such assumptions also yielded a relatively poor agreement (factor of 20 discrepancy) between measured and estimated DNGS Cs-137 coolant activities.

Finally, model predictions are being further validated by comparison with experimental data. For this purpose, measurements of I-129 on spent PHT system purification resins are planned. Also, estimation of the integrated release to date of I-129 from the Darlington PHT system is in progress.

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