MODELLING THE ACTIVITY OF ^{129}I IN THE PRIMARY COOLANT OF A CANDU REACTOR

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ABSTRACT

A mathematical treatment has been developed to describe the activity levels of ^{129}I as a function of time in the primary heat transport system during constant power operation and for a reactor shutdown situation. The model accounts for a release of fission-product iodine from defective fuel rods and tramp uranium contamination on in-core surfaces. The physical transport constants of the model are derived from a coolant activity analysis of the short-lived radioiodine species. An estimate of 3×10^{-9} has been determined for the coolant activity ratio of ^{129}I/^{131}I in a CANDU Nuclear Generating Station (NGS), which is in reasonable agreement with that observed in the primary coolant and for plant test resin columns from Pressurized and Boiling Water Reactor plants. The model has been further applied to a CANDU NGS, by fitting it to the observed short-lived iodine and long-lived cesium data, to yield a coolant activity ratio of ~2×10^{-8} for ^{129}I/^{137}Cs. This ratio can be used to estimate the levels of ^{129}I in reactor waste based on a measurement of the activity of ^{137}Cs.

1. INTRODUCTION

Scaling factors are generally employed to estimate the concentrations of difficult-to-measure (DTM) radionuclides. They relate the activity of DTM radionuclides to the activity of easy-to-measure (ETM) gamma-emitting marker radionuclides such as ^{60}Co and ^{137}Cs.

Due to the much lower concentrations of ^{129}I and ^{99}Tc, their experimental scaling factor development poses some challenges and theoretical treatments have been proposed for their estimation.[1,2,3,4] Such treatments implicitly assume that the scaling factor for radioactive waste can be estimated from coolant activities; departure from this assumption can be empirically accounted for using an experimentally-derived correction factor. The PROFIP code, for instance, has been developed to estimate coolant activity concentrations of fission products and actinides in pressurized water reactors (PWRs).[2] Similarly, the 3R-STAT code focuses on an estimation of ^{129}I and ^{99}Tc coolant activities using measured activities of ^{60}Co, ^{137}Cs and short-lived radioiodine species.[4]

The current work presents the development of a method for predicting the coolant activity of the long-lived ^{129}I, and its scaling factor relative to that of ^{137}Cs, based on the measured activities of the short-lived iodines, namely, ^{131}I, ^{132}I, ^{133}I, ^{134}I and ^{135}I. This prediction is based on a model that considers the physical mechanisms of release for the iodine species from defective CANDU fuel rods and uranium contamination on in-core surfaces. Thus, the application for this work is for the assessment of the ^{129}I activity in waste packages based on ^{137}Cs gamma emission.

2. MODEL DEVELOPMENT

With the occurrence of defective fuel, volatile fission products that are released from the solid fuel matrix into the fuel-to-clad gap can migrate along the gap to the defect site where they may be released into the primary heat transport system (PHTS) (Section 2.1). Fission products are also generated from tramp uranium on in-core piping and fuel bundle surfaces (Section 2.2). The radioiodine species released from either the defective fuel or tramp uranium will be dissolved in the reactor coolant where they can be removed from the PHTS with the operation of coolant cleanup systems (Section 2.3).

2.1 Fission Product Release from Defective Fuel Elements

Fission products can be released from the uranium dioxide fuel into the free void space (fuel-to-clad gap) within the fuel element via solid-state lattice diffusion.

The release rate-to-birth rate ratio (R/B) from the fuel matrix for the radioactive species of interest is given by:[5]

$$\left(\frac{R}{B}\right)_{dif} = 3\left[\frac{1}{\sqrt{\mu}}\coth\sqrt{\mu} - \frac{1}{\mu}\right] \approx \frac{3}{\sqrt{\mu}} \qquad \textbf{(1)}$$

where $\mu = \lambda/D'$, $D' = D/a^2$, λ is the radioactive decay constant (s^{-1}), D is the diffusion coefficient in the fuel matrix ($m^2\ s^{-1}$) and a is the "idealized" fuel grain sphere radius. For the stable isotopes, where $\lambda = 0$ (or $\mu \rightarrow 0$), the R/B ratio at time t is:

$$\left(\frac{R}{B}\right)_{dif} = \left[1 - \frac{6}{\pi^2}\sum_{n=1}^{\infty}\frac{e^{-n^2\pi^2\tau}}{n^2}\right]. \qquad \textbf{(2a)}$$

where $\tau = D't$. The short-time approximation for Eq. (2a) (i.e., for $\tau \leq 0.1$) is:

$$\left(\frac{R}{B}\right)_{dif} = \left[6\sqrt{\frac{\tau}{\pi}} - 3\tau\right] \qquad \textbf{(2b)}$$

2.1.1 Mass Balance in the Fuel-to-Clad Gap

Volatile fission products that are released into the gap will migrate towards the defect site where they may be eventually released into the reactor coolant. This transport can be treated as a first-order rate process, as characterized by a gap escape rate coefficient ν.[6] A mass balance equation can be written for the fission product inventory (N_g) in the fuel-to-clad gap, where on accounting for a source due to a diffusional release from the fuel matrix (R_{dif}) and a loss due to radioactive decay:

$$\frac{dN_g}{dt} = R_{dif} - (\lambda + \nu)N_g \qquad \textbf{(3)}$$

Here the release rate from the defective rod is given by:

$$R_c(t) = \nu N_g(t) \qquad \textbf{(4)}$$

Using Eqs. (1), (3) and (4), the release-to-birth rate ratio for fission product release into the coolant (i.e., during steady-state conditions) is:

$$\frac{R_c}{B} = 3\left(\frac{\nu}{\lambda+\nu}\right)\left[\frac{1}{\sqrt{\mu}}\coth\sqrt{\mu} - \frac{1}{\mu}\right] \approx 3\left(\frac{\nu}{\lambda+\nu}\right)\sqrt{\frac{D'}{\lambda}} \qquad \textbf{(5)}$$

Similarly, for the stable isotopes, using Eq. (2a), gives:

$$\frac{R_c(t)}{B} = \left[\left(1-e^{-\psi\tau}\right) - 6\psi\sum_{n=1}^{\infty}\frac{e^{-\psi\tau} - e^{-n^2\pi^2\tau}}{n^2\pi^2\left(n^2\pi^2-\psi\right)}\right] \qquad \textbf{(6)}$$

where $\psi = \nu/D'$.

2.2 Fission Product Release from Tramp Uranium

Uranium contamination resulting from a previous fuel loss from defective fuel rods or from the fuel manufacturing process can lead to additional activity in the PHTS when this contamination is deposited on in-core surfaces. With defective rods, grain boundary oxidation results when the coolant contacts the solid fuel pellet under the defect site, leading to a washout of the individual fuel grains.[7] The possibility for dissolution of UO_2 near the defect site is expected to be extremely small in an alkaline coolant (pH ~ 10) at a coolant temperature of ~300°C.[8] As such, the fuel debris will be typically in the form of small particles that are roughly spherical, with a diameter comparable to that of the grain radius of ~ 10 μm. As a result of a small particle size, the temperature generated by fission heating is generally too low for diffusion to be an important transport process. As such, the temperature-independent process of recoil now becomes the dominant release mechanisms where:[7]

$$\left(R/B\right)_{rec} = 1 \qquad \textbf{(7)}$$

This result is to be expected since any fission fragment generated in the small fuel particle has a sufficiently long range so that it is instantaneously released into the coolant.

If the fuel particle is deposited on a piping surface, the release into the coolant will be one half of that for a particle suspended in the coolant from symmetry arguments since the fission fragment has sufficient kinetic energy to embed itself into the underlying piping surface such that:

$$\left(R/B\right)_{rec} = \tfrac{1}{2} \qquad \textbf{(8)}$$

2.3 Mass Balance in the Reactor Coolant

The fission product release models developed for defective fuel and tramp uranium contamination can be used to provide a scaling factor for the estimation of the coolant activity of the long-lived isotope, ^{129}I. In particular, by fitting the release model to the measured coolant activity data for the short-lived iodine isotopes (Section 2.3.1), the fitted model parameters can then be used for the prediction of ^{129}I (Section 2.3.2).

2.3.1 Short-lived Iodine Analysis

The fission product inventory in the reactor coolant (N_c) can be determined by a mass balance of the source releases from both defective fuel rod(s) R_c (Eq. (5) or (6)) and uranium contamination R_{rec} (Eq. (8)) and losses due to radioactive decay and coolant purification:

$$\frac{dN_c}{dt} = R_c^t - \left(\lambda + \beta_p\right)N_c \qquad \textbf{(9)}$$

where β_p is a coolant purification rate constant (s^{-1}) and

$$R_c^t(t) = R_c(t) + R_{rec} \tag{10}$$

The total rate of release of a given short-lived iodine isotope into the coolant from x defective fuel rods and tramp uranium follows from Eqs. (5), (8) and (10):

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

where $A = x\left(3\sqrt{D'}F_f\right)$ and $c = F_t / 2$. Here F_t is the fission rate in the tramp uranium (fission s^{-1}) and F_f is the average fission rate per defective rod (fission s^{-1}). The parameter H accounts for precursor-diffusion effects, i.e., except for I-132 (where $H \approx 6$), this factor is the order of unity and can be ignored.[9] Fission product yields y and decay constants for the iodine isotopes of interest are given in Ref. 10.

The model in Eq. (11) can be subsequently equated to a measured $(R/y)_{meas}$ ratio determined from steady-state coolant activity measurements A_c ($= \lambda N_c$) (in Bq) via Eq. (9):

$$\left(\frac{R}{y}\right)_{meas} = \left(\frac{\lambda+\beta_p}{\lambda}\right)\frac{A_c}{y} \tag{12}$$

Thus, a fitting of the model to at least three measured activities of the short-lived iodine species yields the model parameters. In particular, the current treatment provides for a characterization of the defective fuel rods(s) and the amount of uranium contamination. This information is contained in the fitted escape rate coefficient ν and average empirical diffusivity D'(i.e., relevant for x defective fuel rods operating at an average fission rate per defective rod of F_f). The amount of uranium contamination is contained in the parameter c, which implicitly distinguishes the fission rate in the tramp uranium from that in the defective fuel rod(s). These parameters can then be employed in a scaling model to predict the coolant activity of the long-lived ^{129}I as detailed in Section 2.3.2.

2.3.2 *I-129 Coolant Activity Behaviour*

In contrast to the short-lived isotopes, the long-lived ones do not necessarily reach an equilibrium in the reactor coolant. Consequently, the mass balance equation in Eq. (9), for the fission product inventory in the coolant, must be directly integrated (i.e., where $\lambda \sim 0$). Here the source release into the coolant from x defective fuel rods and uranium contamination is described by Eqs. (6), (8) and (10):

$$R_c^t(t) = xF_f y\left[\left(1-e^{-\nu t}\right)+\frac{6\nu}{\pi^2}\sum_{n=1}^{\infty}\frac{e^{-n^2\pi^2 D't}-e^{-\nu t}}{n^2\left(n^2\pi^2 D'-\nu\right)}\right]+cy \tag{13a}$$

Equations (9) and (13a) can be numerically solved for the time dependent coolant activity $A_c(t)$ ($= \lambda N_c(t)$) (in Bq) using a Runge-Kutta technique for a time-variable purification constant. For short times, i.e., $\tau = D't \leq 0.1$, Eq. (13a) does not converge very quickly and, alternatively, can be replaced by the short-time approximation:[10]

$$R_c^t(t) = x3F_f\,y\left[\frac{\left(1-e^{-\psi\tau}\right)}{\psi} - \frac{erf\sqrt{-\psi\tau}}{\sqrt{-\psi}}e^{-\psi\tau} - \tau + 2\sqrt{\frac{\tau}{\pi}}\right] + cy \tag{13b}$$

Moreover, an analytical solution of Eqs. (9) and (13a) is possible assuming a time-averaged coolant purification constant $\overline{\beta}_p$ where, with $A_c(0) = 0$,

$$A_c(t) = \mu x F_f\,y\left\{\frac{1-e^{-\phi\tau}}{\phi} + \left(\frac{e^{-\psi\tau}-e^{-\phi\tau}}{(\psi-\phi)}\right)\frac{3}{\psi}\left[1-\sqrt{\psi}\cot\sqrt{\psi}\right] + 6\psi\sum_{n=1}^{\infty}\left(\frac{e^{-\phi\tau}-e^{-n^2\pi^2\tau}}{n^2\pi^2\left(n^2\pi^2-\psi\right)\left(n^2\pi^2-\phi\right)}\right)\right\} + \lambda cy\left(\frac{1-e^{-\phi\tau}}{\overline{\beta}_p}\right) \tag{14}$$

in which $\psi = v/D'$ and $\phi = \overline{\beta}_p / D'$.

This analysis provides an estimate of the long-lived ^{129}I coolant activity as a function of time while the reactor is operating (i.e., prior to a reactor shutdown event). An expression for the coolant activity after reactor shutdown is detailed in Section 2.3.3.

2.3.3 *Effect of Reactor Shutdown*

When the reactor is shutdown, there is generally a greater burden of iodine activity in the PHTS due to the process of "iodine spiking", where iodine deposited on internal fuel and clad surfaces is dissolved by liquid water.[11] This process can be modeled as an instantaneous one since it occurs quite rapidly (i.e., within several hours). Thus, for an impulsive source of release following shutdown (which is assumed to occur at $t = 0$), the coolant activity $A_c(t)$ at time t is given by:

$$A_c(t) = \left[A_{co} + A_{go}\right]e^{-\beta_p t} \tag{15}$$

where A_{co} and A_{go} are the initial quantities in the coolant and gap at the time of shutdown.

The initial coolant activity A_{co} for the long-lived species follows from the analysis of Section 2.3.2. The quantity A_{go} is the initial gap activity when the reactor is shutdown which can be determined with Eqs. (4) and (6), such that:

$$A_{go} = \left[\frac{\left(1-e^{-\psi\tau}\right)}{\psi} - 6\sum_{n=1}^{\infty}\frac{e^{-\psi\tau}-e^{-n^2\pi^2\tau}}{n^2\pi^2\left(n^2\pi^2-\psi\right)}\right]\mu x F_f\,y \tag{16}$$

The steady-state coolant activity for the short-lived species can be further determined, where on rearranging Eqs. (11) and (12):

$$A_{co} = \left[\left(\frac{v}{\lambda+v}\right)3\sqrt{\frac{D'}{\lambda}}\left(\frac{\lambda}{\lambda+\beta_p}\right)\right]xF_f\,y + c\left(\frac{\lambda}{\lambda+\beta_p}\right)y \tag{17}$$

Similarly, using Eqs. (4) and (5), the available gap activity on shutdown is

$$A_{go} = \left[\left(\frac{\lambda}{\lambda + \nu} \right) 3 \sqrt{\frac{D'}{\lambda}} \right] x F_f y \qquad \textbf{(18)}$$

3. DISCUSSION

A coolant activity of ^{129}I can be estimated for a defect occurrence in the Douglas Point reactor, where ~18 defective fuel rods operated at a linear heat rating of about 40 kW/m (i.e., average fission rate per rod of $F_f = 5.96 \times 10^{14}$ fission s^{-1}) to an average fuel burnup of 110 MWh/kgU.[9] A steady-state analysis yields the model fitting parameters of: $D' = 6.8 \times 10^{-10}$ s^{-1}, $\nu = 1.4 \times 10^{-6}$ s^{-1} and $c = 5.2 \times 10^{13}$ fission s^{-1}. A defect residence time of $t \sim 170$ d is estimated from the given fuel burnup and linear power. Based on a purification flow rate through the I-X columns of 8.65 L s^{-1}, a column efficiency of 99%, and a PHTS volume of 67000 L, a constant purification rate parameter of $\beta_p = 1.28 \times 10^{-4}$ s^{-1} is determined.[9]

The predicted coolant activity with Eq. (14) is shown in Fig. 1. Similarly, the coolant activity can be evaluated via a Runge-Kutta numerical integration of Eqs. (9) and (13a) using a time step of $h = 6$ h. The short-time approximation for the release rate in Eq. (13b) can be alternatively used (i.e., for $\tau = 0.010 \leq 0.1$). All three approaches typically deviate by less than ~1%. As shown in Fig. 1, most of the coolant activity in this particular case is derived from defective fuel operation with a smaller contribution resulting from tramp uranium.

As expected, the coolant activity after ~170 d for ^{129}I (i.e., 390 Bq), is many orders of magnitude smaller than that observed for ^{131}I (i.e., 1.3×10^{11} Bq), yielding a ratio of ~ 3×10^{-9}. This predicted ratio is in agreement with measured activities for ^{129}I/^{131}I in pressurized water reactors (PWRs) shown in Section 3.1 (see Table 1 (last column)), which provides confidence in the model.

Any iodine remaining in the gap from the previous constant period of operation will be rapidly released with water entry into the fuel-to-clad gap on reactor shutdown. This release will result in an additional burden of activity in the coolant. Thus, for a shutdown after ~170 d of operation, the initial coolant activity for ^{129}I at the start of the shutdown would be $A_{co} \sim 387$ Bq (Fig. 1). Thus, from Eq. (16), the amount of iodine that would be quickly released from the gap is $A_{go} \sim 34770$ Bq. This "iodine spike" release is several orders of magnitude greater than that determined while the reactor is operating. A similar "iodine-spiking" phenomenon will also occur for the shorter-lived ^{131}I in accordance with Eqs. (15), (17) and (18).

3.1 Comparison to Other Measured and Predicted ^{129}I Coolant Activities

The Electricité de France (EdF) has obtained actual measured coolant activity data for ^{129}I in several PWR plants (see Table 1).[12] These activities can also be compared to those derived with a scaling from gamma measurements of ^{133}Xe and ^{137}Cs using the PROFIP code. Other measured coolant activities for ^{133}Xe, ^{131}I, ^{134}I and ^{137}Cs are also shown in Table 1. It can be seen that predictions of the long-lived ^{129}I activity, based on a given scaling, may vary by an order of magnitude or so from the corresponding measured value. The measured ^{129}I/^{137}Cs ratio is essentially constant for several plants (i.e., within a factor of ~50).

In addition, the Battelle Pacific Northwest Laboratories designed a mixed-bed test resin sampling program to simulate a scaled-down version of the purification demineralizer system for a nuclear power plant.[3] Test resin columns were installed at 10 PWRs and 7 Boiling Water Reactors (BWRs). The columns were operated with scaled-down flow rates to simulate the cleanup of the reactor coolant. Radioanalytical techniques were used to measure the concentrations of ^{129}I, ^{99}Tc, ^{90}Sr, ^{238}Pu, ^{239}Pu, ^{240}Pu and ^{14}C. Thermal emission mass spectrometry was used for the measurement of ^{129}I. Also, gamma-emitting radionuclides were measured by gamma spectrometry. The available concentration measurements of ^{137}Cs, ^{131}I and ^{129}I, and their respective ratios, for various plant test resin columns are shown in Table 2.[3] These ratios are comparable with those depicted in Table 1.

From a waste characterization perspective, the ratio of ^{129}I/^{137}Cs is of greater interest because this ratio can be readily applied with gamma scanning ^{137}Cs data to estimate ^{129}I levels in waste packages. One can first test the model against actual radionuclide activities of other iodine and cesium isotopes. For instance, as depicted in Table 3 for the Darlington Nuclear Generating Station (DNGS), one can benchmark the model against the measured data of ^{137}Cs, ^{131}I , ^{133}I and ^{135}I, and then use the model to subsequently predict the ratio of ^{129}I/^{137}Cs.

For this analysis, it can be assumed that the diffusivity of cesium is approximately equal to that of iodine, i.e., $D'_{Cesium} \sim D'_{Iodine}$, as observed in high-temperature annealing tests.[13] The coolant purification constant will depend on the respective removal efficiency (ε_p) for these species. Moreover, the removal efficiency for cesium is particularly dependent on the operational age of the resin, in which the retention capability is reduced with time due to exchange with other ions such as Li. For instance, in the Battelle study, the fresh resin samples had a comparable efficiency for cesium and iodine $(\varepsilon_p)_{\text{Cesium}} \sim 0.9\ (\varepsilon_p)_{\text{Iodine}}$; however, this relative efficiency is significantly reduced for the reactor coolant purification resins in PWRs where $(\varepsilon_p)_{\text{Cesium}} \sim 0.2\ (\varepsilon_p)_{\text{Iodine}}$.[3]

The coolant activity data for ^{131}I, ^{133}I and ^{134}I in Table 3 can be fitted to the model of Eqs. (11) and (12), and the coolant activity data for ^{137}Cs can be modeled using Eq. (14). Since recent bundle defect rates for CANDU fuel are typically less than 0.1%, it can be assumed that only a single failure exists ($x = 1$). Thus, a steady-state analysis can be performed for the average of the iodine data in Table 3, based on a coolant mass of 280 Mg and nominal purification rate constant of $\beta_p \sim 4 \times 10^{-5}$ s^{-1} (i.e., ~10 kg s^{-1}). The fitted gap escape rate coefficient is $\nu_I \sim 4.4 \times 10^{-8}$ s^{-1} and the fitted value of A implies an empirical diffusion coefficient of $D' = 4.0 \times 10^{-10}$ s^{-1} (which is consistent with a fuel rod power of ~ 40 kW/m).[9] Unfortunately, it was not known when the defect had actually occurred in reactor. For instance, an element containing a small manufacturing defect typically hydrides within ~ 20 d while a fretting defect, which is caused by debris in the coolant, could occur at any time during its operation. It is also not clear when the defect was identified as well as its post-defect residence time (i.e., if it was prematurely discharged). As such, it can be assumed that the defective bundle was discharged at its nominal discharge burnup of ~180 MWh/kgU, which corresponds at 40 kW/m to a defect residence time of $t \sim 200$ d. The fitted value of c is 1.8×10^{12} fission s^{-1} for the tramp contribution.

Using these fitted parameters, a ^{137}Cs concentration of 0.14 μCi/kg is predicted with Eq. (14) (i.e., in agreement with the measured values in Table 3) if it is assumed that $D'_{\text{Cesium}} \sim D'_{\text{Iodine}}$, $\nu_{\text{Cesium}} \sim 3\ \nu_{\text{Iodine}}$ and $(\varepsilon_p)_{\text{Cesium}} \sim 0.1(\varepsilon_p)_{\text{Iodine}}$ (in accordance with PWR experience).[3] The model therefore predicts a ratio for ^{129}I/^{137}Cs of $(2.6 \times 10^{-9}$ μCi/kg)/(0.14 μCi/kg) $\sim 2 \times 10^{-8}$.

This predicted ratio for ^{129}I/^{137}Cs ratio is comparable to that measured for the PWRs in Tables 1 and 2 (second last columns). This result follows somewhat since the coolant-activity ratio is independent of the number of fuel failures, and is relatively insensitive to the fuel rod power, gap escape rate coefficient and coolant purification constant. The time dependence of the coolant activities for ^{137}Cs, ^{129}I and their ratio are also shown in Table 4 for the assumed values of the gap escape rate coefficient and purification constants (i.e., Case I). The sensitivity of the model to the values of these two parameters is further shown in Table 4, for the case when both species have identical chemical behaviour such that

$\nu_{\mathrm{Cesium}} = \nu_{\mathrm{Iodine}}$ and $(\varepsilon_p)_{\mathrm{Cesium}} = (\varepsilon_p)_{\mathrm{Iodine}}$ (i.e., Case II). Interestingly, in both cases, the $^{129}\mathrm{I}/^{137}\mathrm{Cs}$ ratios are relatively constant with time; these values are also consistent with the observed range of values in Tables 1 and 2 for PWR fuel. The isotopic ratio for these cases depends on the relative values for the purification efficiency and gap escape rates for the two nuclides. There is little or no difference between the isotopic ratios for the tramp uranium contribution and for the total activity (i.e., resulting from both tramp uranium and defective fuel) for each case. In fact, when the diffusivities, gap escape rate coefficients and purification constants for cesium and iodine are identical (so that $\psi_{\mathrm{Cesium}} \sim \psi_{\mathrm{Iodine}}$, $\phi_{\mathrm{Cesium}} \sim \phi_{\mathrm{Iodine}}$ and $(\beta_p)_{\mathrm{Cesium}} \sim (\beta_p)_{\mathrm{Iodine}}$), as in Case II, then Eq. (14) and Table 4 indicate that these ratios should simply be equal to the ratio of the fission product activities in the fuel rod. A similar statement also holds for the spike release. If the coolant activity is derived solely from tramp uranium then the coolant activity ratios will similarly scale with the fission yield and decay constant. The slight variation in the isotopic ratio with time in Case II is simply due to a changing yield with burnup due to Pu-239 production.[10]

The corresponding $^{129}\mathrm{I}/^{137}\mathrm{Cs}$ ratio for Case I due to the spiking of the iodine and cesium on shutdown, can be further evaluated with Eq. (16), as $\sim 6 \times 10^{-7}$. This latter ratio is larger by a factor of ~ 30 than that obtained during constant power operation.

4. CONCLUSIONS

1. A model has been developed to estimate the long-lived $^{129}\mathrm{I}$ coolant activity as a function time during constant reactor operation. The model accounts for fission product release from both defective fuel rods and uranium contamination that may be present on in-core reactor surfaces. The current model is derived from a consideration of the physical release mechanism of diffusion from the fuel matrix and a subsequent release from the gap to the primary coolant by a first-order rate process. In comparison, due to lower temperatures that arise in deposited fuel debris on in-core surfaces, direct recoil is considered as the release process into the primary coolant. Thus, the resulting activity in the PHTS can be evaluated from a mass balance, considering these various sources, and a loss due to coolant cleanup operations. A model has been further developed to predict the $^{129}\mathrm{I}$ activity spike that occurs following reactor shutdown due to an instantaneous release of the available gap inventory (stored as a water-soluble deposit) in the defective fuel rods. This additional activity results in a further iodine burden for the PHTS.

2. A coolant activity analysis of the short-lived iodine species can be used to provide the transport parameters for the $^{129}\mathrm{I}$ model (since the short and long-lived iodine species have the same physiochemical behaviour). Based on a steady-state analysis, this approach yields the empirical diffusion coefficient and gap escape rate coefficient for defective fuel, and the tramp uranium fission rate. A calculation was performed for the Douglas Point NGS, in which there were ~18 defective fuel rods operating at ~40 kW/m. The resulting ratio between the measured $^{131}\mathrm{I}$ coolant activity and the $^{129}\mathrm{I}$ prediction after ~170 d of reactor operation with defective fuel was 3×10^{-9}. This value is in good agreement with observed coolant activity ratios for $^{129}\mathrm{I}/^{131}\mathrm{I}$ in French PWRs (i.e., 4×10^{-10} to 8×10^{-9}), and for US BWRs and PWRs (i.e., 1×10^{-9} to 4×10^{-8}) obtained from test resin column samples in a study by the Battelle Pacific Northwest Laboratories. Model parameters can be derived for cesium by matching the model predictions to measured coolant activity concentrations of $^{137}\mathrm{Cs}$ and short-lived radioiodines. In fact, using these speciation-specific parameters, a predicted coolant activity ratio for $^{129}\mathrm{I}/^{137}\mathrm{Cs}$ of $\sim 2 \times 10^{-8}$ was obtained for the Darlington NGS. Once again, this value is in good agreement with that observed in French and US PWRs for the given isotopic ratio.

For the Douglas Point NGS analysis, with reactor shutdown, the stored gap inventory that is released from the fuel rod results in a ~100-fold iodine spike in the coolant activity level.

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Table 1: ^{129}I Primary Activities and Measured ^{129}I/^{131}I Ratios in French PWRs

Plant	Cycle	Primary Activities (MBq/t)							Measured ^{129}I/^{137}Cs	Measured ^{129}I/^{131}I
		^{133}Xe	^{131}I	^{134}I	^{137}Cs	^{129}I				
						Calculated from		Measured		
						^{133}Xe	^{137}Cs			
GRA3	9	1500	75	2500	45	3.6×10^{-8}	1.4×10^{-6}	6.1×10^{-8}	1×10^{-9}	8×10^{-10}
GRA6	5	1500	70	3000	0.85	4.4×10^{-8}	3.6×10^{-8}	2.9×10^{-8}	3×10^{-8}	4×10^{-10}
CHB1	8	80000	1000	5000	100	6.5×10^{-6}	3.1×10^{-6}	5.0×10^{-6}	5×10^{-8}	5×10^{-9}
CHB2	7	9000	50	700	45	7.2×10^{-7}	1.4×10^{-6}	4.0×10^{-7}	4×10^{-9}	8×10^{-9}
BLA3	9	7000	80	1300	8	5.1×10^{-7}	2.4×10^{-7}	3.0×10^{-7}	4×10^{-8}	4×10^{-9}
BLA4	9	50000	600	4000	150	4.0×10^{-6}	4.7×10^{-6}	1.3×10^{-6}	9×10^{-9}	2×10^{-9}
BUG2	13	1000	100	4000	1	5.8×10^{-8}	3.1×10^{-8}	$<1.5\times10^{-6}$	$<2\times10^{-6}$	$<2\times10^{-8}$
GRA2	12	4000	200	8000	80	1.2×10^{-7}	2.5×10^{-6}	3.2×10^{-7}	4×10^{-9}	2×10^{-9}

Table 2: Cesium and Iodine Isotopic Data for US PWR and BWR Plant Test Resin Columns

Name	Type	Date	Concentration (μCi/g resin)			Ratio	
			^{137}Cs	^{131}I	^{129}I	^{129}I/^{137}Cs	^{129}I/^{131}I
Indian Point Unit 2*	PWR	2/13/90	0.378	53.9	0.697×10^{-7}	1.8×10^{-7}	1.3×10^{-9}
Indian Point Unit 3	PWR	1/21/90	6.93	6.86	2.95×10^{-7}	4.3×10^{-8}	4.3×10^{-8}
Ginna	PWR	8/19/90	0.901	42.4	4.85×10^{-7}	5.4×10^{-7}	1.1×10^{-8}
Braidwood Unit 1	PWR	11/29/90	0.245	30.3	9.52×10^{-8}	3.9×10^{-7}	3.1×10^{-9}
Beaver Valley	PWR	11/9/90	1.84	2.89	2.11×10^{-8}	1.1×10^{-8}	7.3×10^{-9}
Vermont Yankee	BWR	4/12/90	1.78	64.4	3.5×10^{-7}	2.0×10^{-7}	5.4×10^{-9}
WNP-2	BWR	3/21/90	4.72	144	9.83×10^{-7}	2.1×10^{-7}	6.8×10^{-9}

* Average of samples IP-2-1, IP-2-3, IP-2-4, IP-2-5.

Table 3: Comparison Between Radioiodine and Other Key Radionuclide PHTS Concentrations in the Darlington NGS

Radionuclide Concentration (μCi/kg)	Unit 1		Unit 2		Unit 3		Unit 3	
	LM*	LD**	LM	LD	LM	LD	LM	LD
^{60}Co	0.11	2.1	0.15	2.3	0.12	2.2	0.096	2.2
^{137}Cs	0.12	2.2	0.22	3.2	0.16	2.4	0.12	3.1
^{131}I	0.16	2.0	0.48	4.4	0.18	3.0	0.23	2.6
^{133}I	0.96	3.6	4.5	9.6	1.3	5.7	2.1	4.3
^{134}I	6.6	3.8	18	16.3	5.9	10.8	9.4	7.7
Ratio of Key Radionuiclides								
^{60}Co/^{137}Cs	0.86	2.1	0.67	2.0	0.79	2.0	0.79	2.4
^{60}Co/^{134}I	0.017	4.1	.0082	10.0	0.020	8.0	0.011	7.6
^{133}I/^{131}I	7.1	1.9	9.3	2.9	7.2	2.6	9.1	2.3
^{134}I/^{131}I	41	3.1	37	4.7	34	4.7	41	4.2

* LM = Log Mean; ** LD = Log Deviation.

Table 4: Time Dependence of the Total and Tramp Activities in the Coolant for the Darlington NGS*

Case I: $v_{Cesium} = 3\, v_{Iodine}$ and $(\varepsilon_p)_{Cesium} = 0.1\, (\varepsilon_p)_{Iodine}$

Time t (d)	Coolant Activity (µCi/kg)				$^{129}I/^{137}Cs$ Ratio	
	Cs-137		I-129		Tramp U	Total Activity (Defective Fuel + Tramp U)
	Tramp U	Total (Defective Fuel + Tramp U)	Tramp U	Total (Defective Fuel + Tramp U)		
10	0.00195	0.00390	4.91×10^{-11}	7.35×10^{-11}	2.5×10^{-8}	1.9×10^{-8}
20	0.00203	0.00866	5.15×10^{-11}	1.24×10^{-10}	2.5×10^{-8}	1.4×10^{-8}
40	0.00204	0.0214	5.56×10^{-11}	2.72×10^{-10}	2.7×10^{-8}	1.3×10^{-8}
60	0.00205	0.0361	5.90×10^{-11}	4.67×10^{-10}	2.9×10^{-8}	1.3×10^{-8}
80	0.00206	0.0513	6.18×10^{-11}	7.00×10^{-10}	3.0×10^{-8}	1.4×10^{-8}
100	0.00207	0.0665	6.42×10^{-11}	9.62×10^{-10}	3.1×10^{-8}	1.5×10^{-8}
120	0.00208	0.0813	6.64×10^{-11}	1.25×10^{-9}	3.2×10^{-8}	1.5×10^{-8}
140	0.00208	0.0955	6.84×10^{-11}	1.55×10^{-9}	3.3×10^{-8}	1.6×10^{-8}
160	0.00209	0.109	7.02×10^{-11}	1.88×10^{-9}	3.4×10^{-8}	1.7×10^{-8}
180	0.00210	0.122	7.19×10^{-11}	2.21×10^{-9}	3.4×10^{-8}	1.8×10^{-8}
200	0.00210	0.134	7.35×10^{-11}	2.56×10^{-9}	3.5×10^{-8}	1.9×10^{-8}

Case II: $v_{Cesium} = v_{Iodine}$ and $(\varepsilon_p)_{Cesium} = (\varepsilon_p)_{Iodine}$

Time t (d)	Coolant Activity (µCi/kg)				$^{129}I/^{137}Cs$ Ratio	
	Cs-137		I-129		Tramp U	Total Activity (Defective Fuel + Tramp U)
	Tramp U	Total (Defective Fuel + Tramp U)	Tramp U	Total (Defective Fuel + Tramp U)		
10	0.000202	0.000303	4.91×10^{-11}	7.35×10^{-11}	2.4×10^{-7}	2.4×10^{-7}
20	0.000203	0.000489	5.15×10^{-11}	1.24×10^{-10}	2.5×10^{-7}	2.5×10^{-7}
40	0.000204	0.000999	5.56×10^{-11}	2.72×10^{-10}	2.7×10^{-7}	2.7×10^{-7}
60	0.000205	0.00163	5.90×10^{-11}	4.67×10^{-10}	2.9×10^{-7}	2.9×10^{-7}
80	0.000206	0.00234	6.18×10^{-11}	7.00×10^{-10}	3.0×10^{-7}	3.0×10^{-7}
100	0.000207	0.00310	6.42×10^{-11}	9.62×10^{-10}	3.1×10^{-7}	3.1×10^{-7}
120	0.000208	0.00391	6.64×10^{-11}	1.25×10^{-9}	3.2×10^{-7}	3.2×10^{-7}
140	0.000208	0.00474	6.84×10^{-11}	1.55×10^{-9}	3.3×10^{-7}	3.3×10^{-7}
160	0.000209	0.00559	7.02×10^{-11}	1.88×10^{-9}	3.4×10^{-7}	3.4×10^{-7}
180	0.000210	0.00646	7.19×10^{-11}	2.21×10^{-9}	3.4×10^{-7}	3.4×10^{-7}
200	0.000210	0.00733	7.35×10^{-11}	2.56×10^{-9}	3.5×10^{-7}	3.5×10^{-7}

* For both of these cases, $v_l = 4.4 \times 10^{-8}\ s^{-1}$ and $\beta_l = 3.83 \times 10^{-5}\ s^{-1}$ (as detailed in the text)

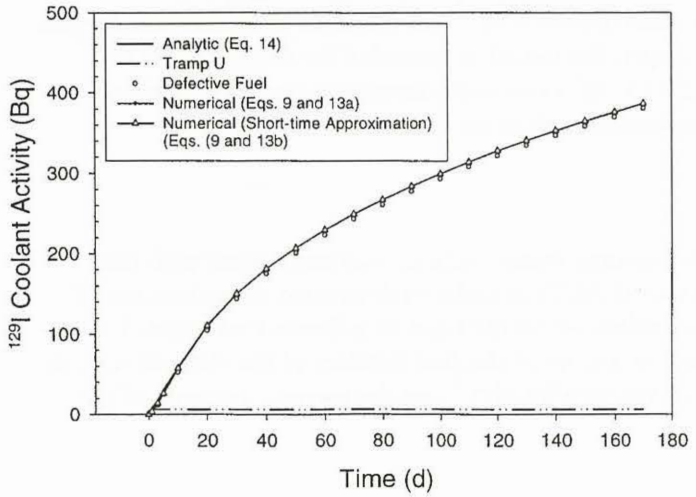

Figure 1. Calculation of the coolant activity for ^{129}I as a function of time based on an analytical solution and using a numerical treatment.