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Modeling the activity of ^{129}I and ^{137}Cs in the primary coolant and CVCS resin of an operating PWR

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Abstract

Mathematical models have been developed to describe the activities of ^{129}I and ^{137}Cs in the primary coolant and resin of the chemical and volume control system (CVCS) during constant power operation in a pressurized water reactor (PWR). The models, which account for the source releases from defective fuel rod(s) and tramp uranium, rely on the contribution of CVCS resin and boron recovery system as a removal process, and differences in behavior for each nuclide. The current models were validated through measured coolant activities of ^{137}Cs. The resultant scaling factors agree reasonably well with the results of the test resin of the coolant and the actual resins from the PWRs of other countries.

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1. Introduction

^{129}I, which is a low energy beta emitter (max energy = 0.15 MeV) with half-life of 1.57×10^7 year, is a critical nuclide for low and intermediate level waste disposal because it is a significant hazardous radionuclide in ground water due to its high mobility in underground water. The inventory of ^{129}I in nuclear waste needs to be accurately analysed before disposal. Due to the difficulty in direct measurement of waste packages, ^{129}I is generally evaluated using a scaling factor between ^{129}I and easier measured ^{137}Cs. The determination of scaling factor is difficult because of the much lower concentration of ^{129}I.

Theoretical approaches have been proposed to estimate ^{129}I activity or scaling factor [1–3]. In these approaches, evaluations of the ^{129}I activity are based on the release of related fission products from the source term, such as defective fuel and tramp uranium. The ratio of the release rate of ^{129}I and ^{137}Cs from the fuel matrix is considered as a constant scaling factor of purification resin or CVCS resin [2,3]. The source term ratio of ^{129}I/^{137}Cs is used for all types of wastes, irrespective of the running conditions of the reactor are [4]. Thus, further consideration was given to the differences in the behaviors of ^{129}I and ^{137}Cs in the primary coolant and in CVCS resin and to the ways these differences affect the scaling factor of ^{129}I.

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Mathematical models have been derived to describe the activities of ^{129}I and ^{137}Cs in the primary coolant and CVCS resin during constant power operation in a PWR. The current model for ^{129}I coolant activity is based on the primary coolant activity model for ^{129}I of a CANDU reactor [5]. The current coolant activity model, especially for ^{137}Cs, can account for the losses caused by a boron recovery system (BRS) at a PWR. Moreover, these models for coolant activity have been expanded for activity in CVCS resin. To distinguish the difference in the behavior of each nuclide, differentiable correlations of the model parameters were applied between ^{129}I and ^{137}Cs. The theoretical scaling factors in the primary coolant and CVCS resin were derived by using the measured reactor coolant system (RCS) data. The resultant scaling factors were compared with the measured results of French and US PWRs.

2. Activity model for ^{129}I and ^{137}Cs in primary coolant and in CVCS resin

The Lewis model [5], which was first introduced for a CANDU-type reactor, can account for diffusional release from fuel matrix and release by recoil from tramp uranium. It is assumed that the release of fission products from defective fuel rods to the coolant is governed by a first-order rate process. The model is based on an averaged behavior of defective fuel rods in a steady-state operation. It can be expressed as an analytic solution for ^{129}I coolant activity.

CVCS resin is the principal means of removing ionic radionuclide from the primary coolant in a PWR. At South Korea's Kori site, for instance, two mixed bed resins and a cation bed resin are used as a CVCS resin. One of the mixed bed resins is in continuous service, which can be supplemented intermittently by the cation bed resin for additional purification. These resins remove ionic fission products, such as anionic iodine and cationic cesium. The removal of the ionic iodine continues throughout the service period of the mixed bed resin due to its large load capacity. However, the removal of the ionic cesium depends highly on the service life of the mixed bed resin due to a relatively faster reduction of load capacity for cations such as Li. In contrast, the cation bed resin principally removes cesium and lithium isotopes. Therefore, the effective removal efficiency of the CVCS resin is kept high for ^{129}I but time-dependent and low for ^{137}Cs.

BRS's main purpose is the recovery of boron. It can also remove ionic nuclides from coolant. Compared with the CVCS resin, BRS removes a relatively small amount of fission products, especially ^{129}I. However, if the load capacity of mixed bed resin for ^{137}Cs is almost lost, the intermittently operating cation bed resin will become the dominant removal mechanism in the CVCS resin. In that case, the amount of ^{137}Cs lost from coolant as an operation of BRS can be comparable to the amount lost as an operation of CVCS resin. Thus, the CVCS resin and the BRS can be considered as a major loss term for ^{137}Cs from the coolant. The current modeling of the coolant activity is extended to the CVCS resin because the CVCS resin is the predominant process for removing fission products, especially ^{129}I.

From the mass balance in the fuel-to-cladding gap, the coolant and the CVCS resin, activities of ^{129}I and ^{137}Cs with time can be directly integrated as a function of time with a constant power operation (see Appendix A).

The coolant activities of ^{129}I and ^{137}Cs are given by

$$A_{\mathrm{c}}(t) = \mu x F_{\mathrm{f}} y \left[\frac{1-\mathrm{e}^{-\phi\tau}}{\phi} + \frac{3}{\psi}\left(\frac{\mathrm{e}^{-\psi\tau}-\mathrm{e}^{-\phi\tau}}{\psi-\phi}\right)\left[1-\sqrt{\psi}\cot\sqrt{\psi}\right] \right.$$
$$\left. +6\psi\sum_{n=1}^{\infty}\frac{\mathrm{e}^{-\phi\tau}-\mathrm{e}^{-n^2\pi^2\tau}}{n^2\pi^2(n^2\pi^2-\psi)(n^2\pi^2-\phi)}\right] + \frac{Cy\mu}{\phi}(1-\mathrm{e}^{\phi\tau}), \tag{1}$$

$$\text{with } \phi = (\beta_{\mathrm{p}}/D') + (\beta_{\mathrm{b}}/D') \quad \text{for } ^{137}\mathrm{Cs}, \tag{2}$$

$$\phi = \beta_{\mathrm{p}}/D' \quad \text{for } ^{129}\mathrm{I}, \tag{3}$$

where $D' = D/a^2$, $\mu = \lambda/D'$, $\tau = D't$, $\psi = v/D'$ and $C = F_t/2$. A_{c} is the primary coolant activity (Bq), λ is the radioactive decay constant (s^{-1}), D is the diffusion coefficient in the fuel matrix (m^2/s), D' is the diffusivity (s^{-1}), a is the radius of the idealized fuel grain sphere (m), t is the fuel residence time (s), x is the number of defective fuel rods, F_f is the fission rate in the defected fuel rods (fission/s) (with the assumption of 200 MeV/fission), F_t is the fission rate in the tramp uranium (fission/s), y is the fission yield (atom/fission), v is the gap escape rate coefficient (s^{-1}), β_p is the coolant purification rate by the CVCS resin (s^{-1}), and β_b is the coolant purification rate by the BRS (s^{-1}).

The activities of ^{129}I and ^{137}Cs in the CVCS resin are given by

$$A_p(t) = \Gamma x F_f y \phi \mu \left[\frac{f(0) - f(\phi)}{\phi} + \frac{3}{\psi} \left(\frac{f(\psi) - f(\phi)}{\psi - \phi} \right) \left[1 - \sqrt{\psi} \cot \sqrt{\psi} \right] + 6\psi \sum_{n=1}^{\infty} \frac{f(\phi) - f(n^2\pi^2)}{n^2\pi^2 (n^2\pi^2 - \psi)(n^2\pi^2 - \phi)} \right] + \Gamma C y \mu (f(0) - f(\phi)), \tag{4}$$

$$\text{with} \quad f(k) = \frac{-(e^{-k\tau} - e^{-\mu\tau})}{k - \mu} \tag{5}$$

$$\text{and } \Gamma = \beta_p/(\beta_p + \beta_b) \quad \text{for } ^{137}\text{Cs}, \tag{6}$$

$$\Gamma = 1 \quad \text{for } ^{129}\text{I}. \tag{7}$$

These analytic solutions were used with model parameters to estimate the activity of fission products. The model parameters were evaluated from the short-lived radioiodine analysis by using Eq. (8) in a steady state condition, and in which the left-hand side is the predicted R/y, $(R/y)_c$, and the right-hand side is the measured R/y, $(R/y)_m$, expressed as follows [5,6]:

$$\left(\frac{v}{\lambda + v} \right) \frac{A}{\sqrt{\lambda}} H + C = \left(\frac{\lambda + \beta_p}{\lambda} \right) \frac{A_{c,SI}}{y}, \tag{8}$$

$$\text{with } y = \frac{(y_0 w \sigma_f)_{U\text{-}235} + (y_0 w \sigma_f)_{Pu\text{-}239}}{(w\sigma_f)_{U\text{-}235} + (w\sigma_f)_{Pu\text{-}239}}, \tag{9}$$

where $A = 3x\sqrt{D'}F_f \cdot F_f$ is the average fission rate per defective fuel rod, H is a precursor correction factor that accounts for the enhanced diffusional release by a relatively long-lived precursor, $A_{c,SI}$ is the measured coolant activity for each short-lived radioiodine (Bq), y_0 is the cumulative fission yield, σ_f is the microscopic thermal fission cross section (5.80×10^{-22} cm^2 for ^{235}U and 7.42×10^{-22} cm^2 for ^{239}Pu at 293.61 K), and w is the specific fissile content (g/kg U) depends on the burnup B (MWD/kg U).

3. Application

The diffusivity of ^{129}I was estimated in the Lewis model from the experimentally derived correlations for CANDU fuel. The model suggests the following parameter relations between ^{129}I and ^{137}Cs for the gap escape rate coefficient, the diffusivity and the removal efficiency in the purification resin at a CANDU reactor: $v_{Cs} = 3v_I$, $D'_{Cs} = D'_I$, $(\varepsilon_p)_{Cs} = 0.1$, and $(\varepsilon_p)_I = 0.99$ [5]. However, different relations were used for the rationalized application to the PWR. To calculate the activities and scaling factors in the primary coolant and the CVCS resin, the following procedures are conducted.

3.1. Short-lived radioiodine analysis

The model parameters (v, A and C) in Eq. (8) were evaluated from regression analyses. The regression method requires information on object function. The object function χ^2, the target of minimization, was defined:

$$\chi^2(\boldsymbol{a}) = \sum_i ((R/y)_{m,i} - (R/y)_{c,i})^2 \quad \text{for } i = {}^{131}I \sim {}^{135}I, \tag{10}$$

where $\boldsymbol{a}$ is a candidate set of model parameters (v, A and C).

For a case study, we took the measured RCS data (Fig. 1), from the bi-weekly geometric mean (GM) of the RCS activities, which cover the period from 2002 to 2004, of the Kori site's unit 1 in a steady state condition [7]. The simplex simulated annealing method and the Levenberg–Marquardt method were used as a regression method [8–10]. The burnup was estimated from the measured ratio of ^{134}Cs/^{137}Cs. The relation was derived for a maximum burnup of 45 MWD/kg U from the reactor physics code ORIGEN-2.2 [11] (Fig. 2).

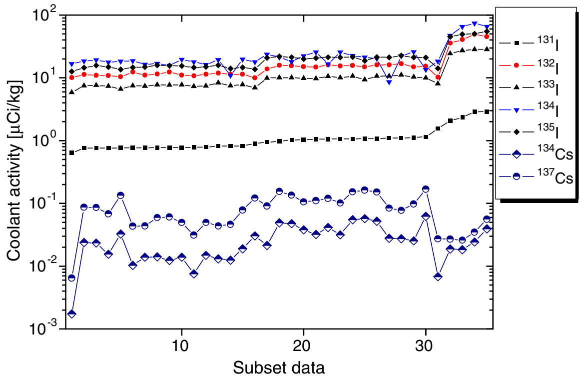

Fig. 1. RCS activities for each subset (order of magnitude of ^{131}I activity).

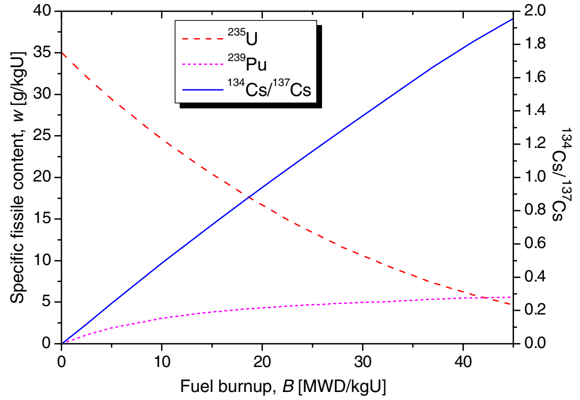

Fig. 2. Specific fissile content and the activity ratio of ^{134}Cs/^{137}Cs versus the burnup in 3.5% enriched fuel.

3.2. Determination of the number of defective fuel rod(s)

To evaluate the D' value for ^{129}I from the derived value of A, the number of defective fuel rods, x, and the average fission rate, F_f must be known. The number of defective fuel rods was postulated from the empirical failure prediction model for the PWR iodine data suggested in the CHIRON code (with the assumption that $x \geqslant 1$) [12]. However, the influence of the resultant value of x is relatively less sensitive to the activity of each nuclide, and the influence to the activity ratio of ^{129}I/^{137}Cs is nearly negligible.

3.3. Diffusivity

The power-related diffusivity of ^{129}I was derived from the Turnbull correlation for the diffusion coefficient, with the assumption that the average oxygen-to-uranium ratio was 2.1 for the defective fuel [13–15]. The corresponding fuel temperature was evaluated from the power-related fuel temperature in Ref. [16]. The diffusivity of ^{137}Cs was estimated by multiplying the derived power-related diffusivity of ^{129}I with the ratio of the cesium-to-iodine diffusivities given as a function of temperature in ANS 5.4 [16]. The resultant power-dependent diffusivities and their ratio are shown in Fig. 3. The diffusivities are taken from the regression analysis as a function of power (P).

Once the number of defective fuel rods is evaluated, the remaining D' and F_f values can be evaluated from the A/x because D' and F_f are given as a function of power. The apparent power is adjusted by increasing the number of defective fuel rods, x, so that the calculated power does not exceed the available maximal power (P_{max}) in the normal operation. Once the corresponding power is calculated, D' and F_f for ^{129}I can be evaluated.

3.4. Gap escape rate coefficient for ^{137}Cs

The iodine released into the gap environment tends to react with one kind of fission product, cesium, and forms stable compound, CsI. Deposited CsI crystals escape via a reaction of ingress water vapor when a defect occurs. In this procedure, the faster transport of iodine than cesium shows that all the iodine is not fettered by fission cesium [17,18]. Thus, it is assumed that the magnitude of v_I is comparable to the magnitude of v_{Cs}, and the difference is not as large as suggested by Lewis. It

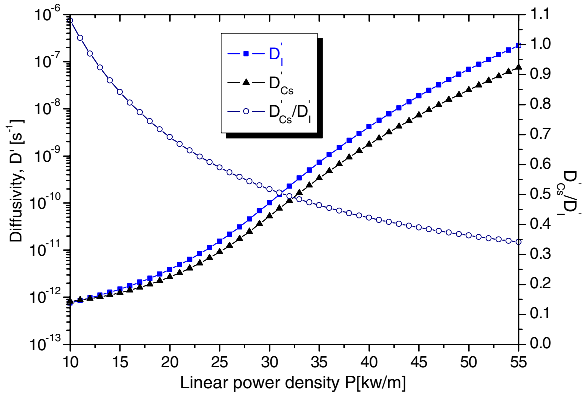

Fig. 3. Diffusivity and the diffusivity ratio as a function of power (with assumption that the value of a is 2.78×10^{-5} m, taken from Ref. [13]).

is assumed that the value of v is identical to each nuclide; that is, $v_I = v_{Cs}$.

3.5. Removal efficiency

Given that the value of ε_p for iodine remains at a high value throughout the entire service period of CVCS resin, it is assumed the value of $(\varepsilon_p)_I$ is 0.99 (that is, DF = 100) [19,20]. However, the total removal efficiency for cesium, ε_{all}, depends highly on the service life of the CVCS resin. Therefore, it can vary widely. To reasonably apply the value of ε_{all}, the optimal ε_{all} was derived by comparing the measured and predicted ^{137}Cs coolant activity on the basis of RCS data (Fig. 1). That is, each optimal ε_{all} was derived by matching the predicted ^{137}Cs activity to the measured ^{137}Cs activity for each subset of the RCS data.

Though the derived ε_{Cs} may include the influence of other factors and other purification mechanisms, the regression result for ε_{all} was used because the contribution of the CVCS resin to the purification is dominant. The value of $(\varepsilon_p)_{Cs}$ was estimated from each value of ε_{all} by using Eq. (A.6) with the assumption that $\varepsilon_b = 0.5$ (corresponding DF = 2) and $\alpha = f_b/f_p = 0.054$. (The value of α was assumed to be four times higher than that in the GALE code because the value of f_b was relatively underestimated. The value of f_b and f_p were given as 0.063 kg/s and 4.662 kg/s, respectively [19].) The resultant removal efficiencies indicate that the contribution of the BRS is negligible when the value of ε_{all} is relatively high (Fig. 4). However, it cannot be negligible when the value of ε_{all} is relatively low. Thus, the resultant correlations are as follows: $v_{Cs} = v_I$; D'_{Cs} and D'_I are given as a different power-related diffusivity, $(\varepsilon_p)_I = 0.99$ and ε_{all} for ^{137}Cs is given as a function of ^{131}I/ ^{137}Cs, and $(\varepsilon_p)_{Cs}$ in Eq. (A.6) (with $\varepsilon_b = 0.5$ and $\alpha = 0.054$).

4. Results and discussion

Model parameters can be evaluated from regression analyses. For example, subset No. 15 results were plotted as R/y versus the decay constant (Fig. 5). The activities of ^{129}I and ^{137}Cs in the coolant and CVCS resin are shown with burnup in Fig. 6; the estimated burnup was 4.97 MWD/kg U. The contributions of tramp uranium to activities in coolant and CVCS resin, which are latter terms in Eqs. (A.11) and (4), respectively, will be generally negligible when meaningful fuel defects exist. The estimated total activity of ^{137}Cs in CVCS resin will be decreased when the effect of BRS is considered, and the difference increases as the value of Γ in Eq. (4) decreases. The relative difference of total activity between coolant and CVCS resin for ^{129}I will be greater than that for ^{137}Cs because the removal efficiency of ^{129}I is generally higher than that of ^{137}Cs in CVCS resin.

RCS data in Fig. 1 was used to compare the predicted and measured ^{137}Cs coolant activities and validate the current models. Different correlations were assumed for comparison: case (i) the existing correlation [5] with D' for ^{129}I in this method; case (ii) a new correlation with GM of the optimal ε_{all}; and case (iii) a new correlation with ε_{all} as a function

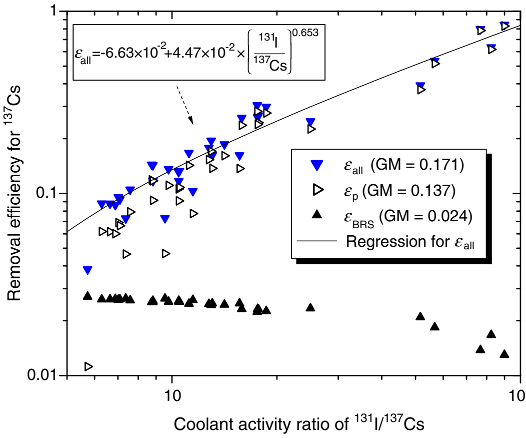

Fig. 4. Removal efficiency for ^{137}Cs versus the activity ratio of ^{131}I/^{137}Cs (the value of ^{131}I/^{137}Cs ranges from 5 to 100 in this case).

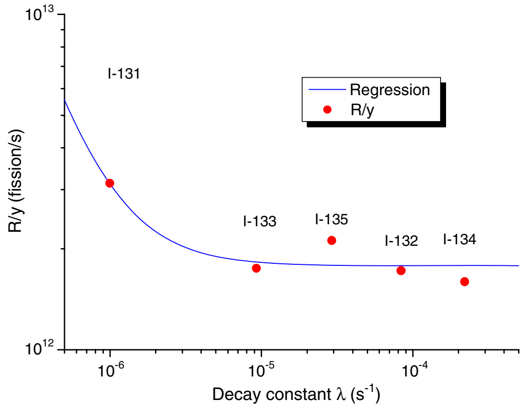

Fig. 5. The R/y versus the decay constant for short-lived radioiodines (with $v = 3.11 \times 10^{-9}$ s^{-1}, $A = 4.32 \times 10^{11}$ fission/s$^{1.5}$, $C = 1.78 \times 10^{12}$ fission/s and $\chi^2 = 1.5347 \times 10^{23}$ fission^2/s^2).

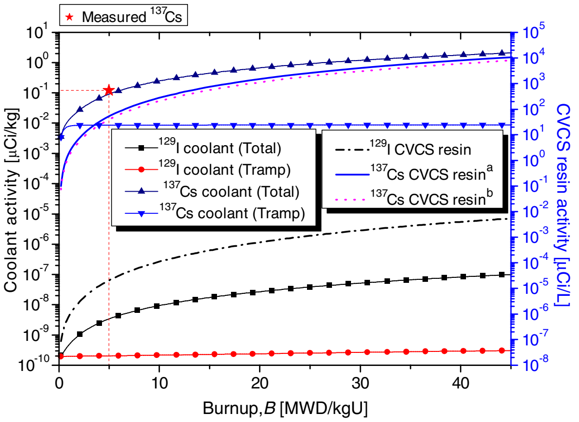

Fig. 6. Coolant and CVCS resin activities for ^{129}I and ^{137}Cs as a function of burnup (the calculated $P = 36.7$ kW/m, $x = 1$; $\varepsilon_{\text{all}} = 0.098$; $(\varepsilon_p)_{\text{Cs}} = 0.073$; $\varepsilon_{\text{BRS}} = 0.025$; the measured and predicted ^{137}Cs coolant activities are 0.122 μCi/kg and 0.090 μCi/kg, respectively (with $M = 1.73 \times 10^5$ kg, $f_p = 2.64$ kg/s and assumed volume of CVCS resin is 1.7×10^3 L in Kori unit 1)) ^{a}: BRS is not considered. ^{b}: BRS is considered.

of ^{131}I/^{137}Cs. The results are summarized in Fig. 7. The mean percentage error (%), PErr(%), which is a measure of the average accuracy of the predictions, is expressed as follows:

$$\mathrm{PErr}(\%) = \frac{1}{n}\sum_{k=1}^{n}\left|\frac{A_{\text{mea}} - A_{\text{pre}}}{A_{\text{mea}}} \times 100\right|,$$
$$k = 1, 2, 3, \ldots, n, \tag{11}$$

where A_{mea} and A_{pre} are the measured and predicted ^{137}Cs coolant activity (μCi/kg), respectively. The present method makes good predictions, and has better results than the previous correlation, especially when the value of ε_{all} as a function of ^{131}I/^{137}Cs was used.

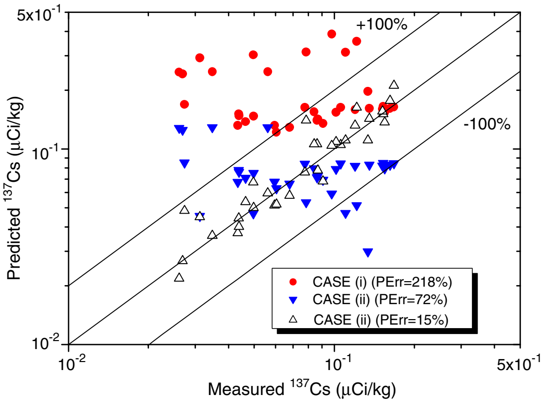

Fig. 7. Comparison of the measured and the predicted ^{137}Cs coolant activity.

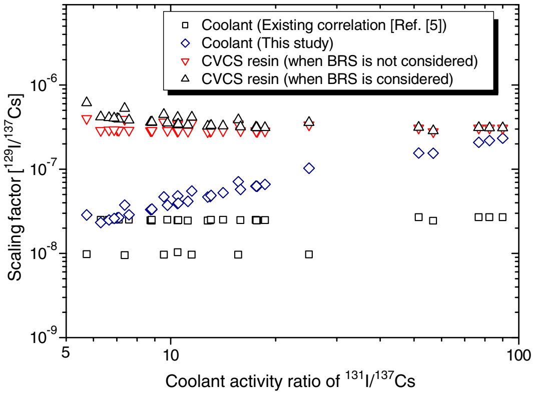

Fig. 8. Comparison of the scaling factors in the coolant and in the CVCS resin.

The resultant scaling factors were plotted by the coolant activity ratio of ^{131}I/^{137}Cs (Fig. 8). The coolant scaling factors, based on the current method with variable ε_{all}, range from 2.32×10^{-8} to 2.33×10^{-7} with GM $= 5.28 \times 10^{-8}$. This range was comparable to, but more conservative than, that of the previous correlation (which ranged from 9.52×10^{-9} to 2.68×10^{-8} and GM $= 2.07 \times 10^{-8}$); in this case the diffusivity for ^{137}Cs derived in this study was used. This result was more variable because the value of ε_{all} was variable. The direct comparison of the resultant scaling factors from results in France and in the US [2,4], are summarized in Table 1. These factors can be affected by parameters such as coolant mass, purification rate, purification system and its operation history, and fuel defect history. Even so, the resultant coolant scaling factors were in good agreement with the results of the test resin in France and in the US.

The scaling factors in CVCS resin were higher than those in coolant. In particular, there was a higher scaling factor in the CVCS resin when BRS was considered (that is, the scaling factor ranges from 2.86×10^{-7} to 6.15×10^{-7} with GM $= 3.61 \times 10^{-7}$) than when BRS was not considered (in which case, it ranges from 2.79×10^{-7} to 3.99×10^{-7} with GM $= 3.03 \times 10^{-7}$). Differences, which ranged from 101% to 154%, depended on the value of Γ. These scaling factors in CVCS resin were comparable to but more conservative than the theoretical source terms scaling factor (3×10^{-7}) [2,4]. Moreover, the scaling factors agree well with the values in the resins generated in French PWRs.

^{137}Cs activity and the scaling factor of ^{129}I in the primary coolant are controlled by the total removal

Table 1
Measured scaling factor in French and US PWRs

Test resin in US PWRs [2]		Test resin in French PWRs [4]		Actual resins in French PWRs [4]	
Plant	$^{129}I/^{137}Cs$	Plant	$^{129}I/^{137}Cs$	Plant	$^{129}I/^{137}Cs$
Oconee 2	7.47×10^{-8}	CHB1	5.00×10^{-8}	GRA5/6	6.3×10^{-7}
Oconee 3	5.76×10^{-8}	CHB2	8.89×10^{-9}		6.5×10^{-9}
Crystal River	1.33×10^{-7}	GRA2	4.00×10^{-9}	CHB1/2	4.7×10^{-9}
Braidwood 2	3.82×10^{-9}	GRA3	1.36×10^{-9}		1.3×10^{-9}
Indian Point 3	4.24×10^{-8}	GRA6	3.41×10^{-8}	BLA3/4	2.2×10^{-6}
Indian Point 2	1.05×10^{-7}	BLA3	3.75×10^{-8}		1.9×10^{-7}
Beaver Valley	1.15×10^{-8}	BLA4	8.67×10^{-9}	BUG2	4.4×10^{-7}
Ginna	5.49×10^{-7}	BUG2	1.50×10^{-6}		1.0×10^{-6}
Oconee 3	1.20×10^{-8}				
Oconee 3	9.20×10^{-9}				
Braidwood 1	3.89×10^{-7}				
Indian Point 2	4.44×10^{-9}				
Byron	1.07×10^{-7}				
Crystal River	1.54×10^{-6}				
GM	5.43×10^{-8}	GM	2.12×10^{-8}	GM	9.08×10^{-8}
AM[a]	2.17×10^{-7}	AM	2.06×10^{-7}	AM	5.59×10^{-7}
SD[b]	4.13×10^{-7}	SD	5.23×10^{-7}	SD	7.53×10^{-7}

[a] Arithmetic mean.
[b] Standard deviation.

efficiency, not by the individual removal efficiency of CVCS resin and BRS. In contrast, ^{137}Cs activity and the scaling factor of ^{129}I in the CVCS resin is influenced by the relative contribution of removal efficiency in the CVCS resin and BRS in relation to total removal efficiency for ^{137}Cs. Thus, the contribution of BRS to the scaling factor in CVCS resin depends on the ratio of the flow rate between BRS and CVCS resin (α), as well as the removal efficiency in CVCS resin for ^{137}Cs ($(\varepsilon_p)_{Cs}$) in our model. As the value of α increases and the value of $(\varepsilon_p)_{Cs}$ decreases, the resultant ^{137}Cs activity in CVCS resin decreases and the resultant scaling factor in CVCS resin increases.

5. Conclusions

1. To theoretically predict the activity of difficult-to-measure ^{129}I, mathematical models were developed to describe the activity of ^{129}I and ^{137}Cs in the primary coolant and in CVCS resin during constant power operation.
2. Having based the models on the release from defective fuel rod(s) and tramp uranium, we considered CVCS resin and BRS as a removal process of fission products from the primary coolant.
3. Total removal efficiency, rather than individual removal efficiency in CVCS and BRS, controls ^{137}Cs activity and the corresponding scaling factor of ^{129}I in primary coolant. In contrast, the contribution of individual removal efficiency in CVCS resin and BRS in relation to total removal efficiency influences ^{137}Cs activity and corresponding scaling factor of ^{129}I in CVCS resin. Therefore, to reliably predict ^{137}Cs activity and scaling factor of ^{129}I in CVCS resin, we must consider how BRS causes a loss of ^{137}Cs from the primary coolant, especially when the load capacity of CVCS resin nearly loses its ion-exchange capacity for cationic ion.
4. RCS data were used to theoretically evaluate the scaling factor. The resultant scaling factors ranged from 2.32×10^{-8} to 2.33×10^{-7} in the coolant. In addition, scaling factors in CVCS resin ranged from 2.79×10^{-7} to 3.99×10^{-7} when BRS was not considered independently, and ranged from 2.86×10^{-7} to 6.15×10^{-7} when BRS was considered independently. These results are in good agreement with radiochemical analysis of RCS sampling in French and US PWRs.

However, the current method is based on the averaged behavior of the defective fuel rods in a steady-state condition. As a result, the level of uncertainty can increase because the defective fuel rods can be affected by power history, the degree of fuel defects, history of coolant cleanup, and related variable reactor history. In addition, uncertainty will increase with spiking of fission products due to shutdown or significant power reduction, though spiking effect can be ignored in long-term steady operation [2,20]. As a result, applied assumptions should be considered in this method. Further work is needed to validate the current method on the basis of domestic sampling of RCS and spent resin.

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Appendix A. Model development

A.1. Mass balance in the primary coolant and CVCS resin

A schematic diagram of the mass balance in the fuel-to-cladding gap, the coolant and CVCS resin is shown in Fig. 9, where CVCS resin is considered a unit system. CVCS resin means a combination of a CVCS mixed bed resin and a CVCS cation bed resin. It is assumed that all fission products removed from primary coolant by CVCS resin were accumulated in the CVCS resin. The fission product inventory in the fuel-to-cladding gap (N_g (atom) in Eq. (A.1)) can be expressed as diffusional release from the fuel matrix, and as losses due to the release from defected fuel rod(s) to the primary coolant and radioactive decay [5]. The fission product inventory in the primary coolant (N_c (atom) in Eq. (A.2)) can be expressed as a mass balance of the source release from defective fuel rod(s) and tramp uranium, and as losses due to the coolant purification by CVCS resin and BRS, respectively, and radioactive decay. The fission product inventory in CVCS resin (N_p (atom) in Eq. (A.5), where p refers to the purification) can be expressed as the accumulated fission products in CVCS resin and loss due to radioactive decay. BRS removes a relatively small amount of ^{129}I, but a large amount of ^{137}Cs in a normal operation. Thus, Eq. (A.2) can be expressed as Eqs. (A.3) and (A.4) for ^{129}I and ^{137}Cs, respectively. The mass balance is written as follows:

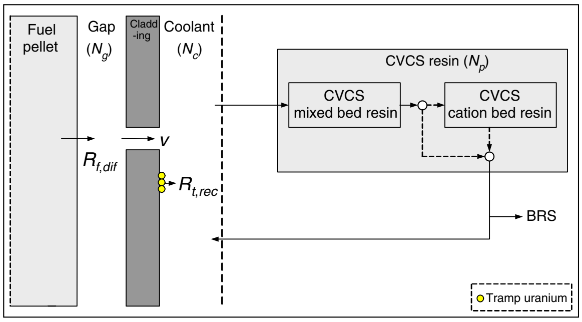

Fig. 9. Schematic diagram of the mass balance.

$$\frac{dN_g}{dt} = R_{f,dif} - vN_g - \lambda N_g \quad \text{(in gap)}, \tag{A.1}$$

$$\frac{dN_c}{dt} = vN_g + R_{t,rec} - \frac{N_c}{M}\varepsilon_p f_p - \frac{N_c}{M}\frac{1}{DF_p}\varepsilon_b f_b - \lambda N_c \quad \text{(in coolant)}, \tag{A.2}$$

$$\frac{dN_c}{dt} \simeq vN_g + R_{t,rec} - \beta_p N_c - \lambda N_c \quad (\text{for } ^{129}\text{I in coolant}), \tag{A.3}$$

$$\frac{dN_c}{dt} = vN_g + R_{t,rec} - \beta_p N_c - \beta_b N_c - \lambda N_c \quad (\text{for } ^{137}\text{Cs in coolant}), \tag{A.4}$$

$$\frac{dN_p}{dt} = \beta_p N_c - \lambda N_p \quad \text{(in CVCS resin)}, \tag{A.5}$$

where $R_{f,dif}$ is the diffusional release rate from the fuel matrix (atom/s), $R_{t,rec}$ is the release rate by recoil from tramp uranium (atom/s), v is the gap escape rate coefficient (s^{-1}), λ is the radioactive decay constant (s^{-1}), $DF_p = 1/(1-\varepsilon_p)$ is the decontamination factor (DF) in CVCS resin, $\beta_b = f_p\varepsilon_p/M$ is the coolant purification rate by the CVCS resin (s^{-1}), $\beta_b = f_p\alpha(1-\varepsilon_p)\varepsilon_b/M$ is the coolant purification rate by BRS (s^{-1}), $\alpha = f_b/f_p$ is the ratio of flow rate between BRS and CVCS resin, f_p is the purification flow rate in CVCS resin (kg/s), f_b is the purification flow rate in BRS (kg/s), ε_p is the removal efficiency in CVCS resin, ε_b is the removal efficiency in BRS, and M is the mass of water in the primary heat transport system (kg). The total removal efficiency, ε_{all}, for ^{137}Cs is defined as a combination of removal efficiencies in CVCS resin and BRS, and is expressed:

$$\varepsilon_{all} = \varepsilon_p + \varepsilon_{BRS} \quad \text{with } \varepsilon_{BRS} = \alpha(1-\varepsilon_p)\varepsilon_b, \tag{A.6}$$

where ε_{BRS} is the effective removal efficiency due to BRS.

The parameters $R_{\mathrm{f,dif}}$ and $R_{\mathrm{t,rec}}$ can be expressed as follows [5,21]:

$$R_{\mathrm{f,dif}} = 3B_{\mathrm{f}}\left[\frac{1}{\sqrt{\mu}}\coth\sqrt{\mu} - \frac{1}{\mu}\right] - 6B_{\mathrm{f}}\mathrm{e}^{-\mu\tau}\sum_{n=1}^{\infty}\left[\frac{\mathrm{e}^{-n^2\pi^2\tau}}{n^2\pi^2+\mu}\right]$$

$$\text{(for long-lived radionuclides),} \qquad (\mathrm{A.7})$$

$$\text{and } R_{\mathrm{f,dif}} = 3B_{\mathrm{f}}\sqrt{\frac{D'}{\lambda}} \quad \text{(for short-lived radionuclides),} \qquad (\mathrm{A.8})$$

$$R_{\mathrm{t,rec}} = \frac{1}{2}B_{\mathrm{t}}, \qquad (\mathrm{A.9})$$

where $B_{\mathrm{f}} = F_{\mathrm{f}}y$, $B_{\mathrm{t}} = F_{\mathrm{t}}y$, $D' = D/a^2$, $\mu = \lambda/D'$, and $\tau = D't$. Here, B_{f} is the birth rate in fuel matrix (atom/s), $F_{\mathrm{f}} = 3.121\times10^{13}\ P\cdot L$ is the fission rate in the defected fuel rods (fission/s) (with the assumption of 200 MeV/fission), P is the linear power density (kw/m), L is the fuel element length (m), y is the fission yield (atom/fission), B_{t} is the birth rate in tramp uranium (atom/s), F_{t} is the fission rate in tramp uranium (fission/s), D is the diffusion coefficient in the fuel matrix (m^2/s), D' is the diffusivity (s^{-1}), a is the radius of the idealized fuel grain sphere (m), and t is the fuel residence time (s).

A.2. Activities of ^{129}I and ^{137}Cs in the primary coolant and in the CVCS resin

Mass balance equations were directly integrated as a function of time with a constant power operation [5]. The activities of ^{129}I and ^{137}Cs in the gap are derived from Eqs. (A.1) and (A.7), and are expressed as Eq. (A.10). The activities of ^{129}I and ^{137}Cs in the coolant are derived from Eqs. (A.3), (A.4), (A.9) and (A.10), and are expressed them in Eqs. (A.11)–(A.13):

$$A_{\mathrm{g}}(t) = \frac{3B_{\mathrm{f}}\mu}{(\mu+\psi)}\left[\frac{1}{\sqrt{\mu}}\coth\sqrt{\mu} - \frac{1}{\mu}\right](1-\mathrm{e}^{-(\mu+\psi)\tau}) - 6\mu B_{\mathrm{f}}\mathrm{e}^{-\mu\tau}\sum_{n=1}^{\infty}\frac{\mathrm{e}^{-\psi\tau}-\mathrm{e}^{-n^2\pi^2\tau^2}}{(n^2\pi^2-\psi)(n^2\pi^2+\mu)}, \qquad (\mathrm{A.10})$$

$$A_{\mathrm{c}}(t) = \mu x F_{\mathrm{f}} y\left[\frac{1-\mathrm{e}^{-\phi\tau}}{\phi} + \frac{3}{\psi}\left(\frac{\mathrm{e}^{-\psi\tau}-\mathrm{e}^{-\phi\tau}}{\psi-\phi}\right)[1-\sqrt{\psi}\cot\sqrt{\psi}] + 6\psi\sum_{n=1}^{\infty}\frac{\mathrm{e}^{-\phi\tau}-\mathrm{e}^{-n^2\pi^2\tau^2}}{n^2\pi^2(n^2\pi^2-\psi)(n^2\pi^2-\phi)}\right] + \frac{Cy\mu}{\phi}(1-\mathrm{e}^{-\phi\tau}), \qquad (\mathrm{A.11})$$

$$\text{with } \phi = (\beta_{\mathrm{p}}/D') + (\beta_{\mathrm{b}}/D') \ \text{ for } ^{137}\mathrm{Cs}, \qquad (\mathrm{A.12})$$

$$\phi = \beta_{\mathrm{p}}/D' \ \text{ for } ^{129}\mathrm{I}, \qquad (\mathrm{A.13})$$

where $\psi = v/D'$ and $C = F_{\mathrm{t}}/2$, with the assumption that $A_{\mathrm{g}}(0) = A_{\mathrm{c}}(0) = 0$ and that $v \gg \lambda$, $\beta_{\mathrm{p}} \gg \lambda$ for volatile long-lived fission products in a typical defective fuel [22]. Here, $A_{\mathrm{g}}(=\boldsymbol{N}_{\mathrm{g}}\lambda)$ is the gap activity (Bq), $A_{\mathrm{c}}(=\boldsymbol{N}_{\mathrm{c}}\lambda)$ is the primary coolant activity (Bq), x is the number of defective fuel rods.

The activity of ^{129}I in CVCS resin, $A_{\mathrm{p}}(=\boldsymbol{N}_{\mathrm{p}}\lambda)$, is derived analytically from Eqs. (A.5) and (A.11) with the assumption that $A_{\mathrm{p}}(0) = 0$, and is expressed in a simplified form with a function $f(k)$ as follows:

$$A_{\mathrm{p}}(t) = \Gamma x F_{\mathrm{f}} y \phi\mu\left[\frac{f(0)-f(\phi)}{\phi} + \frac{3}{\psi}\left(\frac{f(\psi)-f(\phi)}{\psi-\phi}\right)\left[1-\sqrt{\psi}\cot\sqrt{\psi}\right] + 6\psi\sum_{n=1}^{\infty}\frac{f(\phi)-f(n^2\pi^2)}{n^2\pi^2(n^2\pi^2-\psi)(n^2\pi^2-\phi)}\right] + \Gamma C y\mu(f(0)-f(\phi)), \qquad (\mathrm{A.14})$$

$$\text{with } f(k) = \frac{-(\mathrm{e}^{-k\tau}-\mathrm{e}^{-\mu\tau})}{k-\mu} \qquad (\mathrm{A.15})$$

$$\text{and } \Gamma = \beta_{\mathrm{p}}/(\beta_{\mathrm{p}}+\beta_{\mathrm{b}}) \quad \text{for } ^{137}\mathrm{Cs}, \qquad (\mathrm{A.16})$$

$$\Gamma = 1 \quad \text{for } ^{129}\mathrm{I}, \qquad (\mathrm{A.17})$$

where Γ is the correlation factor that accounts for the contribution of CVCS resin to the overall coolant purification by CVCS resin and BRS.

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