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The Prediction Methods of Iodine-129 release rate : Model Development

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Abstract

The results of performance assessment analyses have shown that the long-lived radionuclides such as I-129 control the potential individual dose impact to the public. I-129 is difficult-to-measure(DTM) in low-level waste because it is non-gamma emitting radionuclides and exists at extremely low concentrations in radioactive waste generated by nuclear reactors. In this study, computer modeling technique to predict release rate of I-129 is developed to provide another tools for performance assessment of land disposal facilities and characteristics of radwaste. Model suggested in this study will give conservative values of I-129 release rate for determination of radwaste characteristics. More detailed approach is implemented to account for release conditions of fuel-source-nuclides. I-131 concentration measured from reactor coolant and released fraction from tramp fuel have dominant roles in calculating release rate of I-129 with fuel defect conditions.

1. Introduction

Disposal of radioactive waste requires a license application to contain sufficient information and analyses to provide reasonable assurance that the performance objectives in the regulation will be met. The results of performance assessment analyses have shown that the long-lived radionuclides such as I-129 control the potential individual dose impact to the public[1]. This radionuclides from anionic chemical species in groundwater and consequently their movement through soil is not significantly retarded by interaction with the soil. Due to the long half-live of I-129(*15.7million years*), barriers, geochemical retardation and long travel times will have little or no impact on reducing the individual doses.

I-129 is difficult-to-measure(DTM) in low-level waste because it is non-gamma emitting radionuclides and exists at extremely low concentrations in radioactive waste generated by nuclear reactors. Though results of measurement are taken to describe the characteristics of radwaste, there exist various uncertainties such as laboratory analytical techniques implemented at commercial laboratories or representativeness of the samples. The use of these unrealistic values thus can overestimate the inventories of DTM radionuclide and corresponding ground water dose impact to members of the public by the same amount.

In this study, computer modeling technique to predict release rate of I-129 is developed to provide another tools for performance assessment of land disposal facilities and characteristics of radwaste.

2. Model Development

The basic model assumes steady state condition of reactor and separates release sources of fission products(FP) such as I-129[2]. As for each release sources, it is partitioned considering the FP release mechanisms such as recoil, diffusion and knockout. Basic balance equations of radionuclides in reactor coolant(RC) is derived with steady state conditions:

$$A_i = \frac{\lambda_i y_i}{(3.7E+4) V_{RC} (\lambda_i + \beta)} \left[f(R_i/B_i) + f^T (R_i^T/B_i^T) \right] \qquad \text{[Eq. 1]}$$

where the terms in the brackets represent the release-to-birth(R/B) ratios of defective fuel rods and tramp fuel(T) respectively. To predict release rate of I-129, short-lived iodine periodically measured in RC is utilized to account for release conditions of I-129. Thus, the activity ratio for any pair of iodine isotopes in RC can be derived to account for release conditions of FP as for each release mechanisms such as:

$$\frac{A_i}{A_{I-131}} = \frac{\lambda_i y_i (\lambda_{I-131} + \beta)}{\lambda_{I-131} y_{I-131} (\lambda_i + \beta)} \left[\bar{A} \frac{(R^T/B^T)_i}{(R^T/B^T)_{I-131}} + \bar{B} \frac{(R/B)_i}{(R/B)_{I-131}} \right] \qquad \text{[Eq. 2]}$$

where $\bar{A}$ means release fraction of I-131 from tramp fuel and $\bar{B}$ from defective fuel rods.

For the three fuel release mechanisms, recoil, diffusion, and knockout, the R/B ratios for an isotope pair can be related to the ratio of the decay constants for the iodine isotopes pair as follows:

$$\frac{(R/B)_i}{(R/B)_j} = \left(\frac{\lambda_i}{\lambda_j}\right)^n \qquad \text{[Eq. 3]}$$

The value for n is related to the fuel release mechanism. Each release mechanism have its decay dependence of 0.0, -0.5 and -1.0 for recoil, diffusion and knockout respectively. Radioiodine released within fuel-to-cladding gap has additional decay time before released into RC. This decay constant dependence of the release rates pertains to the defect size in the fuel rod. The activity ratio of any pair of iodine isotopes measured in reactor coolant can now be written in terms of the fractional contribution from the fuel sources and mechanisms as follows:

$$\frac{A_i}{A_{I-131}} = \mathrm{P}_{\mathrm{i,\,I\text{-}131}}\left[\left(\bar{A}q_1+\bar{A}q_2\,h^{-0.5}+\bar{A}q_3\,h^{-0.1}\right)+\left(\bar{B}q_4+\bar{B}q_5\,h^{-0.5}+\bar{B}q_6\,h^{-1}\right)h^{-m}\right] \qquad \text{[Eq. 4]}$$

where

$$P_{i,I-131} = \frac{\lambda_i y_i (\lambda_{I-131}+\beta)}{\lambda_{I-131} y_{I-131} (\lambda_i+\beta)} \qquad \text{[Eq. 5]}$$

$$\bar{B} = 1 - \bar{A} \qquad \text{[Eq. 6]}$$

q_1, q_2 and q_3 mean the release fraction of isotope A_i that is released from tramp fuel by recoil, diffusion and knockout mechanisms and q_4, q_5 and q_6 mean release fraction isotope A_i from fuel contained fuel cladding by recoil, diffusion and knockout mechanisms respectively. The additional exponent, m, is related to the decay time of radionuclide in fuel-to-cladding gap.

For given set of iodine concentrations measured in RC, [Eq. 4] can be solved to give parameters of release condition such as $\bar{A}$, $\bar{B}$, q_1, q_2, q_3, q_4, q_5 ,q_6 and m. To reduce the number of unknowns, the values for tramp fuel such as q_1, q_2 and q_3 are assigned as constant values[2]. There are four iodine pairs(*I-132/I-131, I-133/I-131, I-134/I-131, I-135/I-131*) in the form of [Eq. 4] plus the defined relationships of:

$$\begin{aligned} \bar{A} + \bar{B} &= 1 \\ q_4 + q_5 + q_6 &= 1 \end{aligned} \qquad \text{[Eq. 7]}$$

For a total of six equations that form the basic equations, they are solved simultaneously in this study. When the core and fuel release conditions have been determined, the release rate of I-129 are calculated. These release rates are determined by multiplying the release rate of I-131, due to each mechanism and each fuel source, by appropriate activity release rate ratios as:

$$\begin{aligned} R_{I-129} &= \left(\frac{R_{I-129}}{R_{I-131}}\right)_r R_{I-131,tramp,r} + \left(\frac{R_{I-129}}{R_{I-131}}\right)_d R_{I-131,tramp,d} + \left(\frac{R_{I-129}}{R_{I-131}}\right)_k R_{I-131,tramp,k} \\ &= \left(\frac{R_{I-129}}{R_{I-131}}\right)_r R_{I-131,fuel,r} + \left(\frac{R_{I-129}}{R_{I-131}}\right)_d R_{I-131,fuel,d} + \left(\frac{R_{I-129}}{R_{I-131}}\right)_k R_{I-131,fuel,k} \end{aligned} \qquad \text{[Eq. 8]}$$

$R_{I\text{-}131,\ tramp,[r,d,k]}$ represents the release rates of I-131 from tramp fuel due to each mechanism and is related with fuel release condition calculated from [Eq. 4] such as:

$$R_{I-131,tramp,r} = \bar{A} R_{I-131} q_1 \qquad \text{[Eq. 9]}$$

$$R_{I-131,tramp,d} = \bar{A} R_{I-131} q_2 \qquad \text{[Eq. 10]}$$

$$R_{I-131,tramp,k} = \bar{A} R_{I-131} q_3 \qquad \text{[Eq. 11]}$$

$$R_{I-131} = A_{I-131} V_{RC} (\lambda_{I-131} + \beta) \qquad \text{[Eq. 12]}$$

and for defective fuel rods as:

$$R_{I-131,fuel,r} = \bar{B} R_{I-131} q_4 (\varepsilon + \lambda_{I-131}) / \varepsilon \qquad \text{[Eq. 13]}$$

$$R_{I-131,fuel,d} = \bar{B} R_{I-131} q_5 (\varepsilon + \lambda_{I-131}) / \varepsilon \qquad \text{[Eq. 14]}$$

$$R_{I-131,fuel,k} = \bar{B} R_{I-131} q_6 (\varepsilon + \lambda_{I-131}) / \varepsilon \qquad \text{[Eq. 15]}$$

where ε represent fuel rod escape rate coefficients[4]

3. Result and Discussion

Real RC data of Kori NPP are utilized to show the calculation results. Fig[1] shows the input characteristics of iodine measured from RC and calculated results of I-129 release rate. I-129 may release relatively small amount compared to other iodine and show wide fluctuations during reactor cycle. To have reasonable release rate of I-129, this fluctuation must be removed through average scheme.

Fuel release conditions of iodine is partitioned as intended in Fig[2]. For release conditions of tramp fuel, each portions of release mechanism are calculated by analytic equations[2] and calculated release portion of fuel rod have similar values with those of tramp fuel. There are wide fluctuations in their partition value. Though this model show wide fluctuations in example calculation, averaged release rate of I-129 will give conservative range for prediction value. Concentration of I-131 have dominant effects upon prediction value and the parameter that is related with defect size of fuel rod(m) gives important effects simultaneously. Fig[3] shows I-131 release fractions from tramp fuel($\bar{A}$) and defect size parameter(m).

4. Conclusion

Model suggested in this study will give conservative values of I-129 release rate for determination of radwaste characteristics, if removed their wide fluctuation and reasonably averaged. More detailed approach is implemented to account for release conditions of fuel-source-nuclides. I-131 concentration measured from RC and released fraction from tramp fuel have dominant roles in calculating release rate of I-129 with fuel defect conditions.

It is thought that precursor effects of iodine may reduce relatively wide fluctuations and give more realistic result of its release rate

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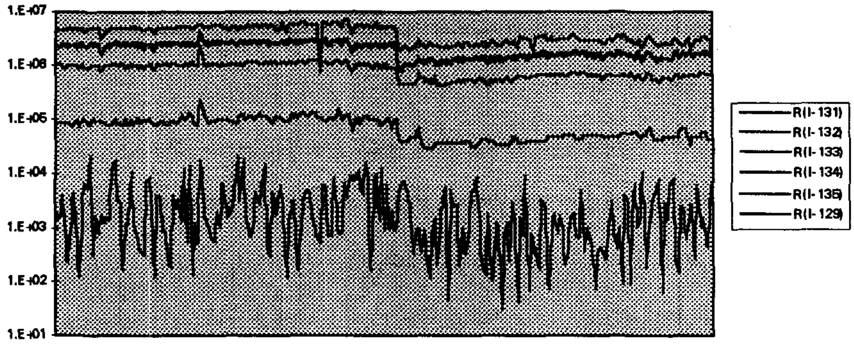

Fig. 1. Input characteristics of iodine calculated results of I-129 release rate

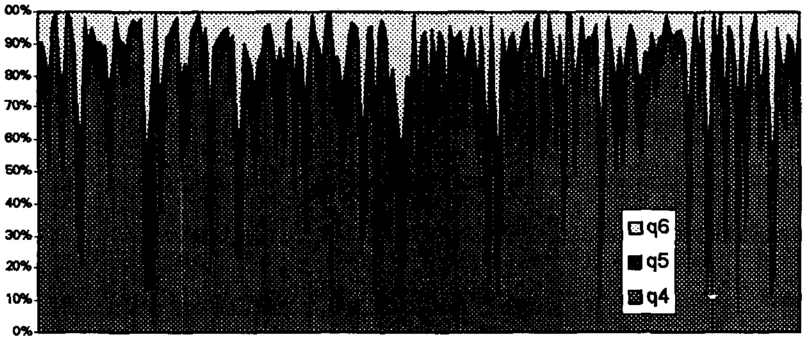

Fig. 2 Fuel release conditions of iodine[Defective fuel rod]

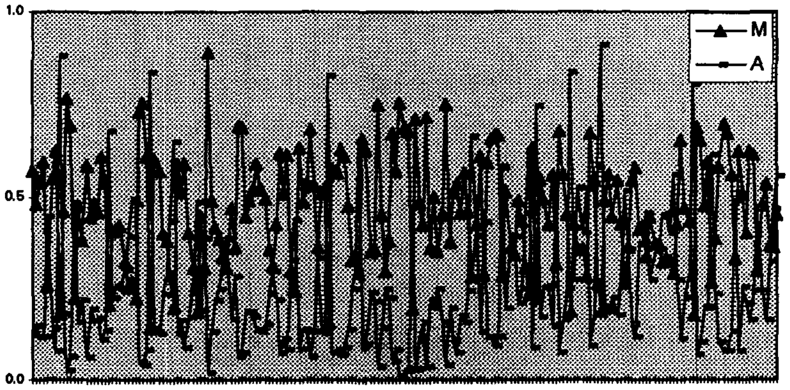

Fig.3 I-131 release fractions from tramp fuel($\overline{A}$) and defect size parameter(m).