

**Development of a Scaling Factor Prediction Method
for Radionuclide Composition in Low-level Radioactive Waste**

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Abstract

This study presents a method to predict plant-specific and operational history dependent scaling factors. Realistic and detailed approaches are taken to find scaling factors at reactor coolant. This approach begins with fission product release mechanisms and fundamental release properties of fuel-source nuclide such as fission product and transuranic nuclide.

Scaling factors at various waste streams are derived from the predicted reactor coolant scaling factors with the aid of radionuclide retention and build up model. This model makes use of radioactive material balance within the radioactive waste processing systems. According to input parameters of plant operation history, scaling factors predicted at reactor coolant and waste streams are well brought out the effects of plant operation history.

1. Introduction

An environmentally sound and safe management of low-level radioactive waste require the knowledge of the nature and the quantities of radionuclides in the immobilized or packaged waste. Accordingly, regulations and guides developed in various countries of the world required detailed description of the waste package and its contents(i.e 10CFR61). The determination of composition of radioactive waste is associated with a number of problem. Often, representative sampling is not possible or results in considerable radiation exposure to sampling personnel. Further, many of radionuclides required for shipping manifests emit no gamma radiation and must be analyzed by complicated radiochemical procedures.

Previous studies [1,2,3] are about statistical scaling factors. Statistical scaling factors are made by sampling data that are gathered from off-site laboratories. These statistical scaling factors give somewhat generic and passive tendency toward Nuclear Power Plant(NPP). Accordingly, there exist many disadvantages such as:

- (1) Generic scaling factors can not account for operation history of NPP in detail.
- (2) Sampling procedures must be conducted following substantial change of NPP

Therefore, prediction method of scaling factors that can account for plant-operation history will be suggested in this study. Active approach will start a course of study from theories about fission product release mechanisms to predict reactor coolant scaling factors. A method to find scaling factors at waste stream will be also suggested. There are many nuclides of our interest for safe radioactive waste management. Among these nuclides, this study will treat fuel-source nuclides such as Fission Product(FP) and TRansUranic (TRU) nuclides.

2. Scaling Factors at Reactor Coolant(RC)

Overall structure of this method starts from FPs release theory of tramp fuel and input parameters of plant operation history. Plant-specific and time-dependent input parameters are chosen to give detailed

information of NPP as below. These input parameters must be obtained easily through plant routine programs of RC sampling such as: (1) iodine concentrations, (2) key nuclides concentration of scaling factors(Co-60 or Cs-137), (3) RC letdown (purification) flow rate.

Final equations used to predict RC scaling factors are as follows for FP and TRU nuclides:

$$SF\left(\frac{FP}{KEY}\right)_{RC} = \frac{R_{I-131}}{R_{KEY}} \Big|_{RC} \times \bar{A} \times C_{ko} \times \left[\frac{C_{FP}}{C_{I-131}} \right]_{ORIGENPC-2} + \frac{R_{I-131}}{R_{KEY}} \Big|_{RC} \times \bar{A} \times (1 - C_{ko}) \times \frac{\lambda_{FP} y_{FP}}{\lambda_{I-131} y_{I-131}}, \quad (1)$$

$$SF\left(\frac{TRU}{KEY}\right)_{RC} = \frac{R_{I-131}}{R_{KEY}} \Big|_{RC} \times \bar{A} \times C_{ko} \times \left[\frac{C_{TRU}}{C_{I-131}} \right]_{ORIGENPC-2} \quad (2)$$

where R_i means release rate of isotopes i , \bar{A} means fractional release of I-131 from tramp fuel, C_{ko} means fractional contribution of I-131 released from tramp fuel by knockout mechanism and $[C_i/C_{I-131}]_{ORIGENPC-2}$ means curie inventory ratio of DTM nuclides to I-131 calculated by ORIGENPC-2 code[4].

Release fraction of I-131 from tramp fuel(\bar{A}) is derived from the mass balance of iodine concentrations in RC. Since release rate is directly proportion to radionuclides formation rates, this equation can be written in terms of fractional release (R/B), that is release rate normalized by nuclides birth rate as below:

$$A_i = \frac{\lambda_i y_i}{(3.7E+4)V_{RC}(\lambda_i + \beta)} \left[f(R_i/B_i) + f^T(R_i^T/B_i^T) \right], \quad (3)$$

Activity ratios of any iodine isotopes to iodine-131 and definition of \bar{A} can be shown as below:

$$\frac{A_i}{A_{I-131}} = \frac{\lambda_i y_i (\lambda_{I-131} + \beta)}{\lambda_{I-131} y_{I-131} (\lambda_i + \beta)} \left[(1 - \bar{A}) \frac{(R/B)_i}{(R/B)_{I-131}} + \bar{A} \frac{(R^T/B^T)_i}{(R^T/B^T)_{I-131}} \right], \quad \bar{A} = \frac{f^T(R^T/B^T)_{I-131}}{f^T(R^T/B^T)_{I-131} + f(R/B)_{I-131}} \quad (4)$$

Since R/B is related with decay constant according to the appropriate release mechanisms, ratios of R/B are also related with decay constant. Hence, the activity ratio for any pair of iodine isotopes in RC is shown as:

$$\frac{A_i}{A_{I-131}} = R_{I-131} \left[(1 - \bar{A}) \left(\frac{\lambda_i}{\lambda_{I-131}} \right)^{-n} + \bar{A} \right], \quad P_{i,I-131} = \frac{\lambda_i y_i (\lambda_{I-131} + \beta)}{\lambda_{I-131} y_{I-131} (\lambda_i + \beta)} \quad (5)$$

There are two unknowns in Eq.(5). The one is exponent, n , that has its range from 0.5 to 1.5 according to fuel rod defect conditions, the other is I-131 fractional release from tramp fuel, \bar{A} , that is of our interest.

Knockout constant(C_{ko}) means I-131 fractional contribution of knockout mechanism released from tramp fuel. It is calculated separately by application of fuel-pellet related equations to tramp fuel. The curie inventory ratios for FP/I-131 and TRU/I-131 are taken from ORIGENPC-2 runs for PWR fuel. Fig.1 shows the calculated ratios for FPs and Fig.2 for TRU nuclides based on 33, 000 MWD/MTU. It must be known the fuel burnup to calculate curie inventory ratios in tramp fuel. Specific nuclide pair must be selected to determine the burn up of tramp fuel according to following selection criteria such as:

- 1) they have relatively long half-lives not to reach equilibrium condition in reactor internals,
- 2) nuclide pairs have similar behavior after release process into the reactor coolant.

It is proposed that the release ratios for TRU nuclide will be equal to curie ratios in the tramp fuel and TRU nuclides are released only from tramp fuel and only by a knockout mechanism. It can be found some TRU data source in [2,3] by which the ratios for TRU pair(Pu-238/Pu-239) is selected and

calculated.

3. Scaling Factor at Waste Stream

Scaling factors at RC cleanup system are more important than those of RC. Basic rationale for the development of appropriate scaling factors at waste stream is that all of the radioactive materials ending up in specific waste streams originates in the reactor core and are transported to these waste streams by RC. Radioactive material balances through each waste system are taken into account how scaling factors change from the RC to the various waste systems. Although there is no typical coolant cleanup system, Fig 3, 4 shows, for example, simplified schematic diagrams of a plausible PWR configuration. For CVCS, cleanup component such as primary purification filter(1), primary purification resin(2), and CVCS evaporator bottom(3) in Fig 3 are of our interest.

The quantity of isotope on each components is given as a function of build-up rate(R_i), process run time(t) and isotope decay constant:

$$Q_i = \frac{R_i}{\lambda_i} (1 - e^{-\lambda_i t}) \quad (6)$$

True scaling factors for each component with scaling factors at RC would be as below:

$$S_{1,i} = \frac{Q_{1,i}}{Q_{1,Key}} = S_{\text{reactor}} \frac{DF_{1,Key} (DF_{1,i} - 1) \lambda_{Key} (1 - e^{-\lambda_i t})}{DF_{1,i} (DF_{1,Key} - 1) \lambda_i (1 - e^{-\lambda_{Key} t})} \quad (7)$$

$$S_{2,i} = \frac{Q_{2,i}}{Q_{2,Key}} = S_{\text{reactor}} \frac{DF_{1,Key} DF_{2,Key} (DF_{2,i} - 1) \lambda_{Key} (1 - e^{-\lambda_i t})}{DF_{1,i} DF_{2,i} (DF_{2,Key} - 1) \lambda_i (1 - e^{-\lambda_{Key} t})} \quad (8)$$

$$S_{3,i} = \frac{Q_{3,i}}{Q_{3,Key}} = S_{\text{reactor}} \frac{DF_{1,Key} DF_{2,Key} DF_{3,Key} (DF_{3,i} - 1) \lambda_{Key} (1 - e^{-\lambda_i t})}{DF_{1,i} DF_{2,i} DF_{3,i} (DF_{3,Key} - 1) \lambda_i (1 - e^{-\lambda_{Key} t})} \quad (9)$$

where $S_{\text{reactor},i}$ means scaling factors at RC and $DF_{1,i}$, $DF_{2,i}$ and $DF_{3,i}$ mean decontamination factor(DF) at component 1, 2, and 3 for each isotope.

For PWR radwaste system, for example, similar calculation procedures can be employed as those conducted at primary system as Fig.3. The original source of the radioactive materials that reach the each waste system is reactor coolant leakage. Thus, there is reason to presume that this leakage should have about the same isotopic ratios as the coolant itself. With this assumption, the rate of buildup on the filter and evaporator located at clean and dirty waste in Fig. 4 would be:

$$R_{f,i} = C_i V F_d \left(1 - \frac{1}{DF_{f,i}}\right), \quad (10)$$

$$R_{e,i} = C_i V F_d \frac{1}{DF_{f,i}} \left(1 - \frac{1}{DF_{e,i}}\right). \quad (11)$$

True scaling factors on the filter and evaporator bottom in Fig.4 with scaling factors at RC would be as below and scaling factors for other component in PWR radwaste systems can be calculated with similar procedure:

$$S_f = S_{\text{reactor}} \frac{DF_{f,Key} (DF_{f,i} - 1) \lambda_{key} (1 - e^{-\lambda_i t})}{DF_{f,i} (DF_{f,Key} - 1) \lambda_i (1 - e^{-\lambda_{key} t})} \quad (12)$$

$$S_e = S_{\text{reactor}} \frac{DF_{f,\text{Key}} DF_{e,\text{Key}} (DF_{e,i} - 1) \lambda_{\text{key}} (1 - e^{-\lambda_i t})}{DF_{f,i} DF_{e,i} (DF_{e,\text{Key}} - 1) \lambda_i (1 - e^{-\lambda_{\text{key}} t})} \quad (13)$$

4. Demonstration

Plant-specific and time-dependent operation history sample parameters are analyzed to account for how much these input parameters will affect scaling factors in RC and at waste streams. Characteristics of sample input parameters are analyzed in Fig.5. Concentrations of key nuclides have some fluctuations with increasing tendency for Co-60 nuclide and decreasing tendency for Cs-137 nuclides. Fig.6 shows variation of I-131 release fraction from tramp fuel, $\bar{\alpha}$.

Fig.7 and Fig.8 shows operation history dependent RC scaling factors with Cs-137 and Co-60 respectively. Plant operation history Input parameters give composite influences into RC scaling factor. Among input parameters, concentrations of measured key nuclides have their strong effects on RC scaling factors than other ones. The RC scaling factors with same key nuclides show similar tendency as that of key nuclides. FPs with relatively long half-lives such as Tc-99 (2.12×10^5 years) and I-129 (1.57×10^7 years) do not show large effects on their scaling factors in RC from key nuclides.

The method for scaling factors at waste stream is applied to Korean Nuclear power plant Units 7 and 8 (KNU7&8). There are mainly two terms to contribute the variations of scaling factors through each waste processing systems. The one is decontamination term in the function of DF factors and the other is decay term in the function of waste collection, processing and discharge times. Decay contributions of scaling factors are relatively minor when compared with that of DF contributions at each processing system such as filter, demineralizer or evaporator. If DF values that are used to calculate scaling factors at waste stream are same at specific processing system, there is no variations when compared with the scaling factors at RC. In real calculations, DF values for each isotope and processing times are quoted from AIF/NESP[5] and PWR GALE[6].

Fig. 9 shows, for example, scaling factors at BRS evaporator condensate demineralizer. Time varying scaling factors of LRS evaporator concentrate tank according to input parameters are shown in Fig.10. Scaling factors at waste system are show similar tendency of variation with that of RC. This is due to scaling factors at waste stream depend very seriously on DF values.

References

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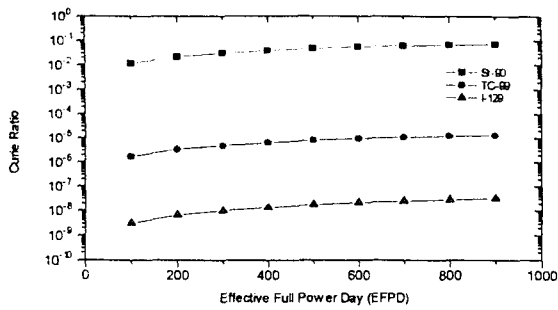


Figure 1 Curie Inventory Ratios for FPs (ORIGENPC-2)

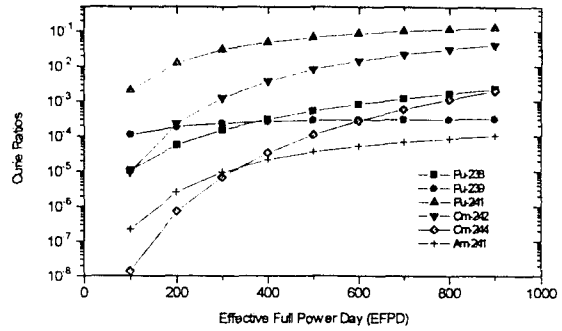


Figure 2 Curie Inventory Ratios for TRU (ORIGENPC-2)

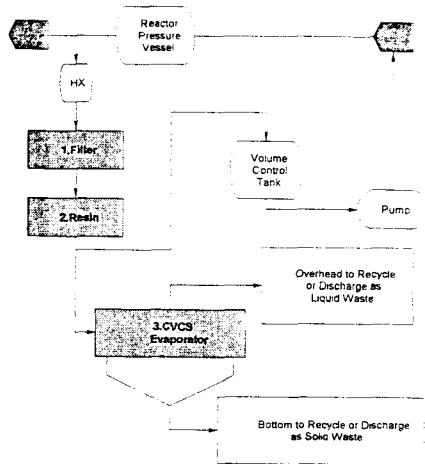


Figure 3 Simplified PWR Primary Cleanup System (CVCS)

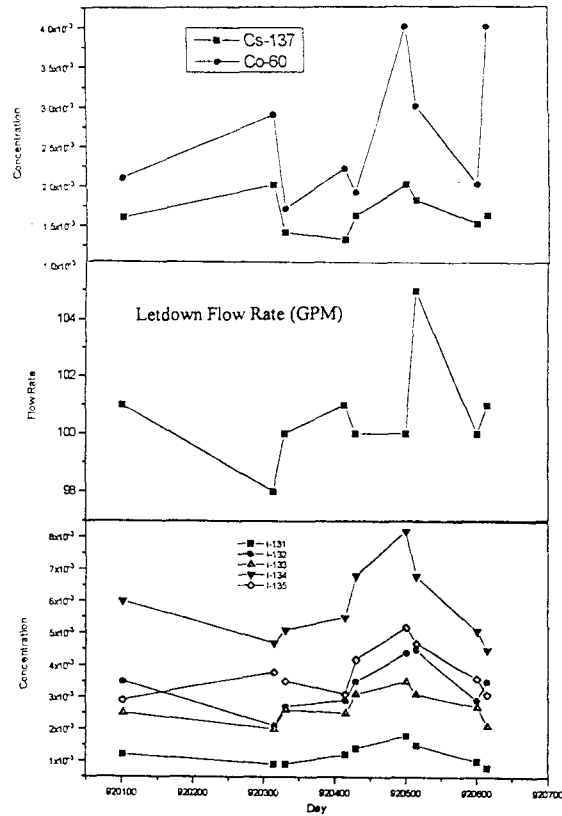


Figure 5 Characteristics of input parameters

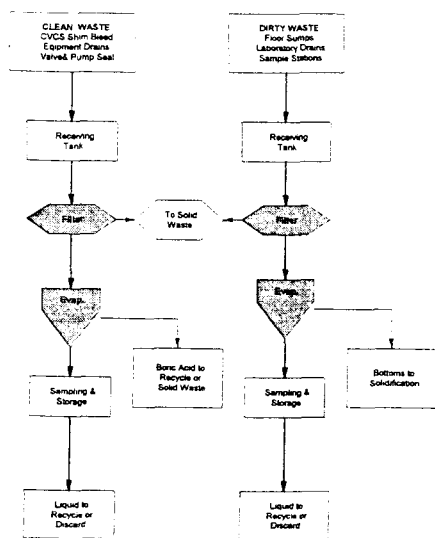


Figure 4 PWR Radwaste Systems (Clean & Dirty Waste System)

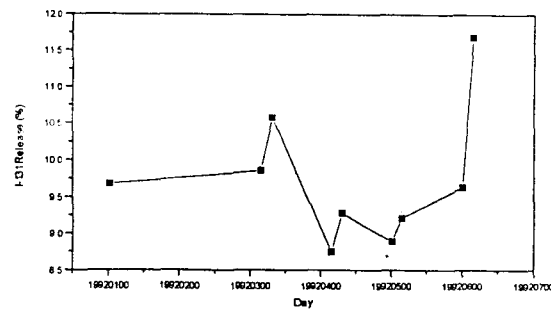


Figure 6 I-131 Release Fractions from Tramp Fuel, \bar{A}

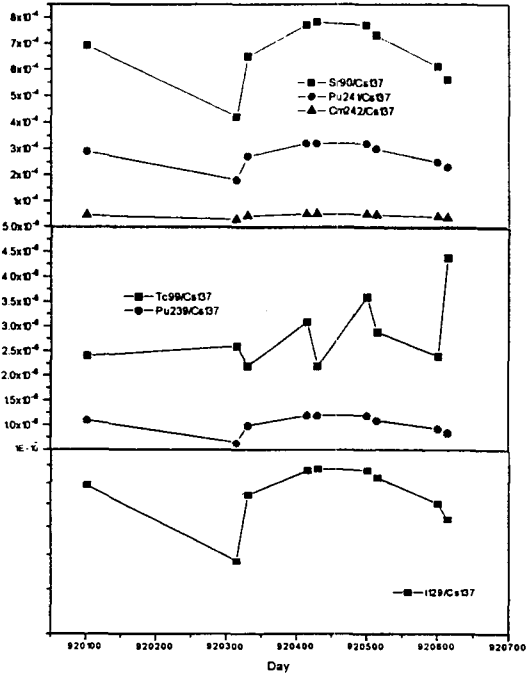


Figure 7 Time Varying SFs in RC (DTM Nuclide/Cs-137)

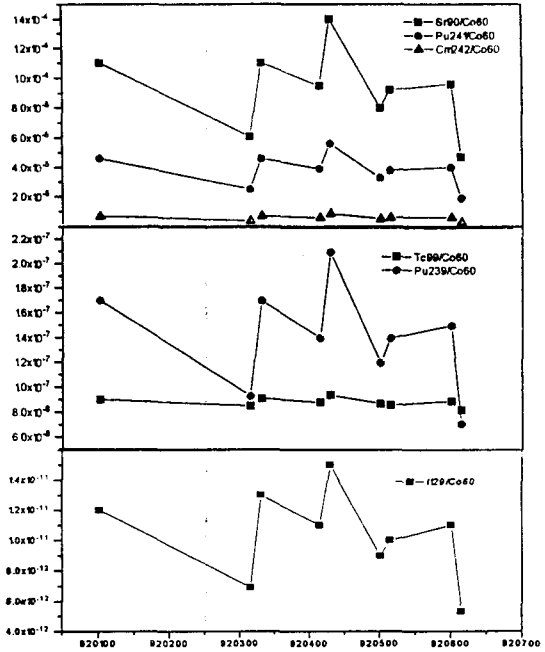


Figure 8 Time Varying SFs in RC (DTM Nuclides/Co-60)

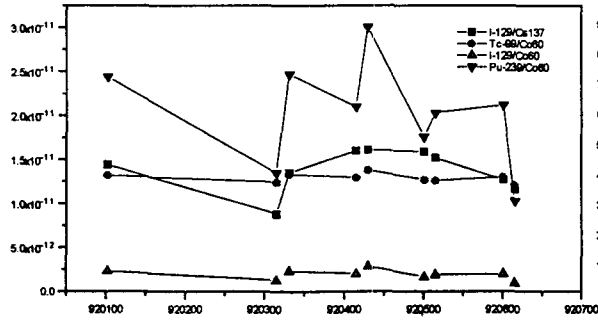
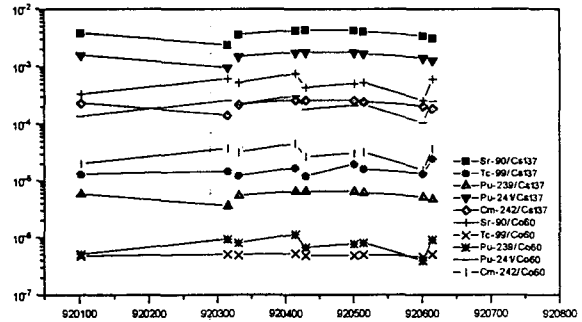
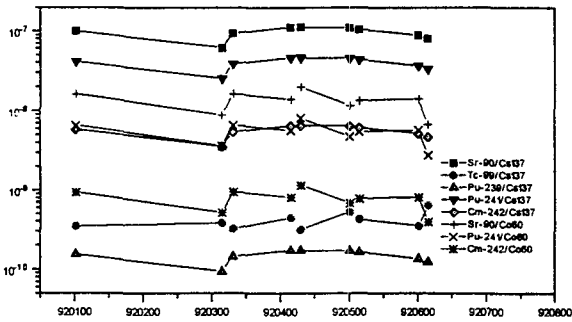


Figure 9 SFs at BRS Evapo. Condensate Demineralizer

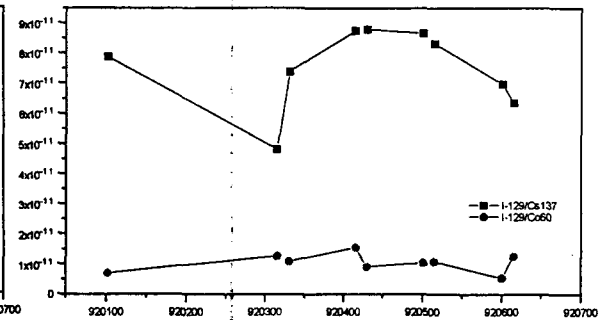


Figure 10 SFs at LRS Evapo. Concentrate Tank