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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

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

$$\text{with } \phi = (\beta_p/D') + (\beta_b/D') \quad \text{for } ^{137}\text{Cs}, \quad (2)$$

$$\phi = \beta_p/D' \quad \text{for } ^{129}\text{I}, \quad (3)$$

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 dif-