EVALUATION OF CRUD BEHAVIOR IN PRIMARY CIRCUIT OF INTEGRAL REACTOR BY MULTI-REGION MODEL

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Abstract

In order to evaluate the behavior of a deposited crud from structual materials surfaces, multi-region model dealing with radioactive corrosion products behavior has been developed to describe the crud behavior in the primary circuit of an integral reactor. The primary circuit of the integral reactor, which adopted ammoniated water chemistry, was divided into nine control volumes. The transportation of the radioactive nuclei by the sedimentation, washing-out, and release of corrosion products were considered between the control volumes and its associated surfaces in the model. Specific activities of the separate radionuclides in the primary coolant as well as on the surfaces of structual materials that were washed by the coolant throughout fuel cycle were also simulated with operation mode. As a result, this multi-regional model was capable of evaluating the behavior of crud more exactly than the two or three nodes model simualtion codes. From this evaluation the integral reactor showed lower activities than the other commercial reactors because it used the titanium alloy for the SG tube material and the avoidance of using boric acid.

1. INTRODUCTION

The reactor coolant system of a pressurized water reactor (PWR) nuclear plant exhibits radioactivity from various sources. These radiation sources consist of the fission products from fuel element and activated products from corrosion products.[1,2] Some of the early commercial nuclear power plants experienced a radiation exposure due to activated corrosion products which was small compared to the radiation from fission products leaking from defective fuel rods. As the design technology and the quality control of a fuel fabrication have improved considerably in the last few decades, the major source of radiation has shifted from the fission products to the corrosion products.[1~3]

The predominant radionuclide in the PWR primary coolant is Co-60, although Co-58 contributes significantly to radioactivity at the early stage of a reactor operation. Since Co-60 is produced from the in-core Co-58, which is the source of cobalt in a reactor system, many researchers have investigated its transmutation in the core, its transport around the system, and its subsequent deposition on surfaces. The major contributor influencing the behavior of radioactivity, such as Co-60 in the coolant, is the corrosion product oxides.[1~4, 7] These oxides originate from the circuit materials in contact with the coolant of the primary circuit, these materials are mainly a zirconium alloy for the core, a titanium alloy for the tubing in the steam generator, and a stainless steel for the reactor vessel internals. The material and water chemistry are selected carefully to control the corrosion products. The integral type reactor developed by KAERI is a soluble boron free reactor, and ammonia is chosen as a pH-reagent.[5,6]

The need to simulate the behavior of corrosion products in a nuclear power plant environment has attracted many researchers' attention. Thus, we have performed the in-depth reviews of the various theories and physics associated with the corrosion products transportation, the laboratory loop testing, and the field measurements in operating plants.

The objective of this work is to analyze the corrosion product activity in the primary circuit of an integral reactor using a multi-region model. This model, which is incorporated with a mass and activity transport between the dissolved corrosion products in the coolant and the surface, describes the behavior of corrosion products in a coolant and on the surfaces according to the operation modes.

2. MULTI-REGION MODEL

This program is an engineering tool for analysis of corrosion products activity in te primary circuits of nuclear power plants. Since there are too many unknowns to predict the activity and radiation level accurately, a simplified model is formulated for this program. This model is aimed at evaluating the corrosion products concentrations in coolant and on the surface of major equipments in the reactor coolant system. This model includes a mass transfer between the various locations of the plant and also accounts for reactor plant operation modes.

The breakdown of the primary circuit is shown in **Fig. 1**. The primary circuit is divided into nine control volumes. Between the control volume and its associated surfaces, the radioactive nuclei are transported by a sedimentation, washing-out, and release of corrosion products. The coolant between the control volumes transports the nuclei, too. Dividing the primary circuit into several regions requires more differential equations to be solved and takes a longer computation time.

During a normal operation, it is assumed that the mass transfer constants are equal for the active and inert corrosion products and that the concentrations of the parent nuclides and activation products in the reactor coolant are in equilibrium. The concentrations are calculated under the consideration of a production rate and removal rate. The parent nuclides are produced by the corrosion of surface materials consisting of the primary circuit and are transported by the coolant into the reactor core region. The parent nuclides are removed by a neutron induced activation in the reactor core region, by a deposition on surface of the primary circuit equipments, or by a purification through the purification system.

The change of parent and active nuclei number per time unit in the i-th system element can be described by the following equations:

$$\frac{dN_i^M}{dt} = \sum_{j=1}^{n} \lambda_{j\to i}(t_k) \cdot N_j^M + W_i^M(t_k) - N_i^M \sum_{j=1}^{n} \lambda_{i\to j}(t_k) \qquad (1)$$

$$\frac{dN_i^D}{dt} = \sum_{j=1}^{n} \lambda_{j\to i}(t_k) \cdot N_j^D + W_i^D(t_k) + + N_i^M \cdot F_{i,l}(E, t_k) - N_i^D(\lambda + \sum_{j=1}^{n} \lambda_{i\to j}(t_k)) \qquad (2)$$

where, N_i^M, N_i^D - number of parent and daughter nuclei in the i-th system element(nucl,); $\lambda_{i\to j}(t_k)$- communication constant of the i-th and j-th system elements in the k-th mode(t_k - mode duration)(1/s) ; $W_i^M(t_k)$ - rate of a parent nuclei entry in the i-th water volume due to surface corrosion(nucl./s) ; $W_i^M(t_k)=\sum_l \rho_l^M \cdot S_l \cdot C_{i,l}(t_k)/\gamma_l$- rate of a parent nuclei entry in the i-th water volume due to a surface corrosion(nucl./s) ; ρ_l^M -parent nuclei concentration in the surface material(nucl/cm^3) ; S_l - surface area washed by the i-th water volume(m^2) ; $C_{i,l}(t_k)$ - rate of corrosion products release in the i-th water volume from surface(S_l) in the k-th operating mode is determined as corrosion rate product by the proportion of corrosion product release to coolant, (g/m^2·s) ; γ_l- surface material density(g/cm^3) ; $W_i^D(t_k)=\sum_l \rho_l^D \cdot S_l \cdot C_{i,l}(t_k)/\gamma_l$ - rate of daughter nuclei release in the k-th mode to the i-th water volume due to surface(S_l) corrosion in the irradiation zone(cm^3/s) ; ρ_l^D- daughter nuclei concentration in the surface(S_l) material (nucl/cm^3).

The change of the parent nuclei concentration in the surface material can be described by the following equation.

$$\frac{d\rho_l^D}{dt} = \rho_l^M \cdot F_{i,l}(E,t_k) - \rho_l^D \cdot \lambda \qquad (3)$$

where, $F_{i,j}(E,t_k)$ - rate of a parent nuclei transition into a daughter nuclei in the i-th element and in the surface material due to irradiation in the k-th mode(s^{-1});

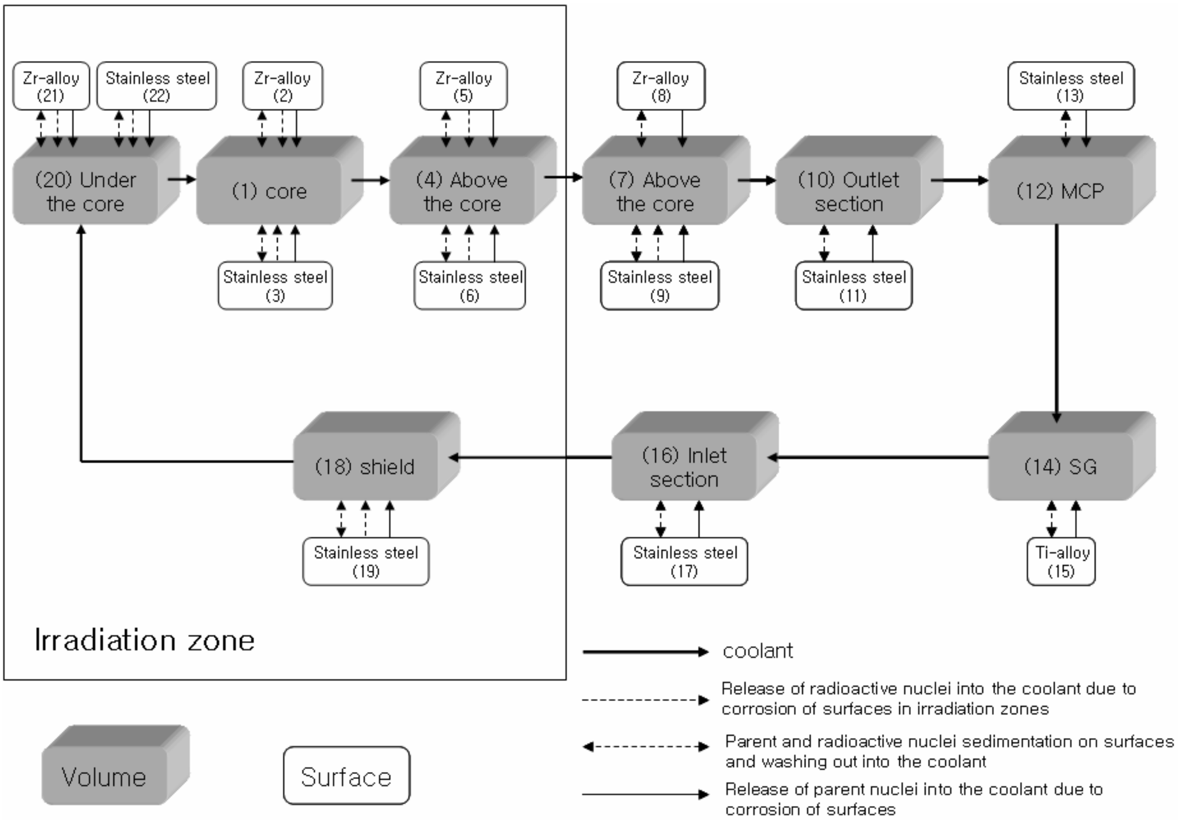


Fig. 1. Breakdown of a reactor plant primary circuit.

For (n-γ)-reactions,

$$F_{i,l}(E,t_k) = \sigma_{thn} \cdot \Phi^i_{thn}(t_k) + \sigma_{res} \cdot \Phi^i_{\succ thn}(t_k)/18{,}2 \qquad (4)$$

For threshold activation reactions;

$$F_{i,l}(E,t_k) = \sigma(E \succ E_{thresh}) \cdot \Phi^i(E \succ E_{thresh}, t_k) \qquad (5)$$

where, $\Phi^i_{thn}(t_k), \Phi^i_{\succ thn}(t_k), \Phi^i(E \succ E_{thresh}, t_k)$ - average density of thermal and epithermal neutron fluxes for the i-th element and surface S_l, and neutron fluxes with energy exceeding E_{thresh} in the k-th operating mode (1/s·cm^2) ; $\sigma_{thn}, \sigma_{res}, \sigma(E \succ E_{thresh})$ - microscopic sections of parent nuclei activation by thermal neutrons, resonance activation integral, and activation section for threshold reactions(cm^2) ; $N_i^M \cdot F_{i,l}(E,t_k)$ - rate of parent nuclei generation in the i-th element of irradiation zone in k-th mode due to parent nuclei activation(nucl./s); λ - decay constant of the formed daughter nucleus(s^{-1}); n – number of elements in the system.

The equation (1) describes parent and daughter nuclei entry in the i-th element of the system from the other elements due to sedimentation, washing out, and transfer with coolant. The equation (2) describes nuclei release from the i-th element of the system to the other elements due to the same processes. Here, the radioactive decay is also taken into account for the daughter nuclei.

Specific activity of corrosion products and concentration of parent nuclei (impurities) at various circuit sections can be determined from the equations (6) and (7):

$$A_i = \lambda \cdot N_i^D / V_i ; \qquad (6)$$

$$C_i = N_i^M / V_i , \qquad (7)$$

where, V_i - geometric characteristic of the element (volume or surface), cm^3 (cm^2)

Description of primary circuit elements and their parameters use in the analyses, chemical composition of surface materials washed by primary coolant are calculated for this program input data.[10] The rates of corrosion products were obtained on the basis of available experimental data on corrosion rate of steels, zirconium alloy and titanium alloy under the ammoniated water chemistry of primary coolant.[8,11,12] The release rates of corrosion products into the coolant were determined as a portion of total corrosion rate. Similar to the release rate of corrosion products into the coolant, the constants of sedimentation and washing out are key parameters for calculation of corrosion activity distribution in the reactor plant process circuit by this program.[11]

Core average activating neutron flux densities are equal to 5.5×10^{13} (cm^{-2}s^{-1}) for thermal neutrons and 6.5×10^{14} (cm^{-2}s^{-1}) for higher than thermal neutrons. Similar values for the reflector are equal to 6.1×10^{13} (cm^{-2}s^{-1}) and 1.3×10^{14} (cm^{-2}s^{-1}), respectively. Of the chemical composition of the structural materials used in this program, stainless steel, titanium alloy, and zirconium alloy are listed in **Table 1**. The sedimentation coefficients of corrosion products on a zirconium alloy and a stainless steel surface are assumed as 6.0×10^{-4} s^{-1} and titanium alloy as 1.0×10^{-4} s^{-1}, respectively. The washing-out constant for all corrosion products and for all surfaces is taken equally to be 5.0×10^{-8} s^{-1}, based on the published data.[8,10]

Table 1. Chemical Composition of Various Materials in Primary Circuit.

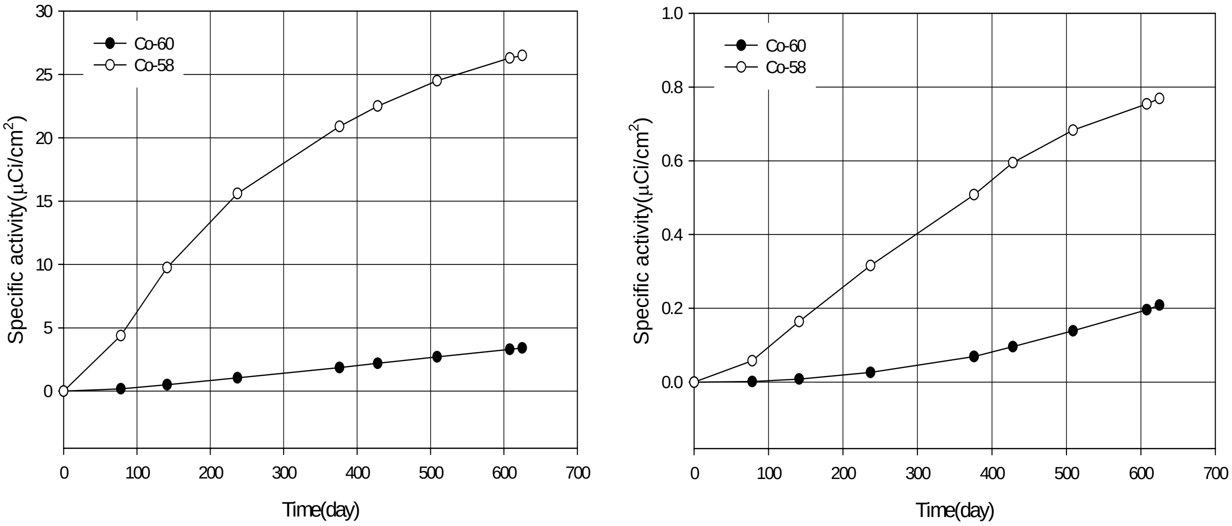
	Material			
	Stainless steel	Inconel	Zr-alloy	Ti-alloy
Fe	67.90	5	0.24	0.25
C	0.08	0.1	-	0.1
Ti	0.70	0.4	-	Main
Zr	-	-	Main	2.0~3.0
Cr	18.00	20~23	-	-
Ni	10.50	58	-	-
Mn	2.00	0.5	-	-
Co	-	1	-	-
Nb	-	3.15~4.15	0.9~1.1	-
Mo	-	8~10	-	-
Al	-	0.4	-	1.8~2.5

3. RESULTS & DISCUSSION

The calculation was done for one fuel cycle of 15,000 hours but the release of the corrosion products into the water was calculated for the time of a metal exposure in water of 24,000 hours. The calculated specific activities of the various species on the surface of te core and steam generator during the effective full power hours, 15,000 hours, are shown in **Fig. 2 (a)** and **(b)**, respectively. On the reactor core surface, the final specific activity of Co-58 was estimated to be 26.5 $\mu Ci/cm^2$. This is 30 times greater than that of the steam generator. The final specific activity of Co-60 on the reactor core surface was 3.41 $\mu Ci/cm^2$, which is 16 times higher than that of the steam generator. It means that a greater part of the generated corrosion product in the reactor was deposited on the core surface. The specific coolant activities of Co-58 and Co-60 in te reactor core were predicted to be 1.4×10^{-3} $\mu Ci/cm^3$ and 2.1×10^{-4} $\mu Ci/cm^3$ as shown in **Fig. 3**. There was no difference in the coolant specific activities between the reactor core and the steam generator.

To compare with the integral reactor, the behaviour of the various radioactive corrosion products was also evaluated for an imaginary reactor (PLANT-I) which has the same configuration as the integral reactor except for the steam generator material and water chemistry. As for the Korea Standard Nuclear Power Plant (KSNP), PLANT-I is assumed to use Inconel-600 for a steam generator material and H_3BO_3-LiOH for a water chemistry control. For the corrosion release rate of the Inconel-600, the published data were used[9] The simulated specific activities of the various species on the surface of the core and steam generator during the effective full power hours, 15,000 hours, are also shown in **Fig. 4 (a)** and **(b)**, respectively. The specific activity of Co-58 on the core surface is also greater than that of the steam generator as shown in the integral reactor. Furthermore, the hydrogen, which was used to remove the dissolved oxygen in the coolant, is not added to the primary coolant. The hydrogen concentration is maintained by ammonia dosing in the primary coolant where hydrogen and nitrogen are generated by a subsequent ammonia radiolytical and thermal decomposition. Thus, the ammonia water chemistry prevents a corrosion of the structural materials contacting the primary coolant during a power operation.

The reactor coolant specific activity of Co-58 in the core is estimated to be 8.32×10^{-2} μCi/cm^3 as shown in **Fig. 5**, which is about 60 times higher than that of the integral reactor. The specific activity of Co-60 in the coolant also shows the big differences in the core and steam generator with a factor of 580. It is considered that the higher values of specific activities of Co-58 and Co-60 in PLANT-I is due to the higher release rate of Inconel, which has the high nickel content of about 60%. The major parameter for this big difference in the specific activities seems to be both the water chemistry and the structural material. As assumed for PLANT-I, the water chemistry in a KSNP is a H_3BO_3-LiOH chemistry. The lithium hydroxide alkali, which is injected for a reaction control in the primary coolant circuit of KSNP to compensate for the acid properties of boric acid, brings about the stress corrosion cracking of steam generator tube and the corrosion of zirconium alloy for a fuel cladding. Furthermore, the hydrogen concentration is maintained by an ammonia dosing in the primary coolant where hydrogen and nitrogen are generated by an ammonia subsequent radiolytical and thermal decomposition.[11,12] Thus, the ammonia water chemistry prevents a corrosion of the structural materials contacting the primary coolant during power operation.



(a) **(b)**

Fig. 2 Specific activities according to time (day) on the surface of (a) core and (b) steam generator of the integral reactor.

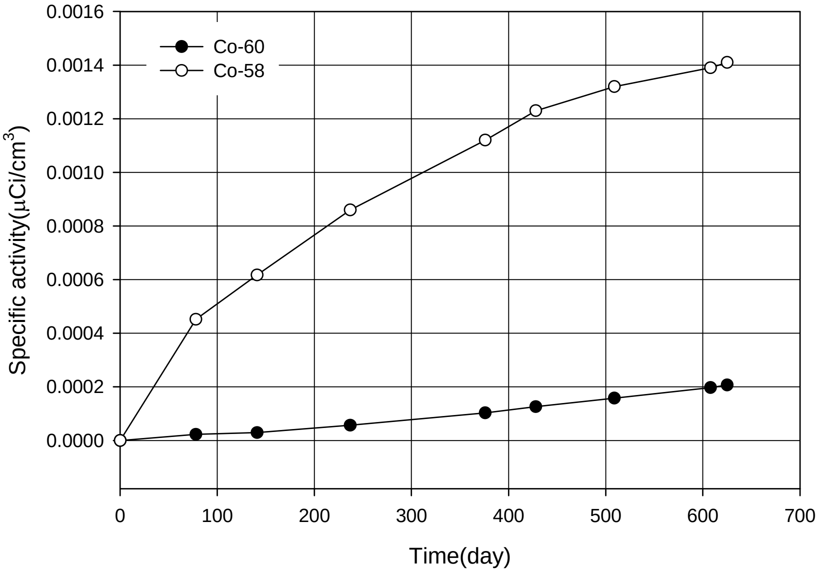
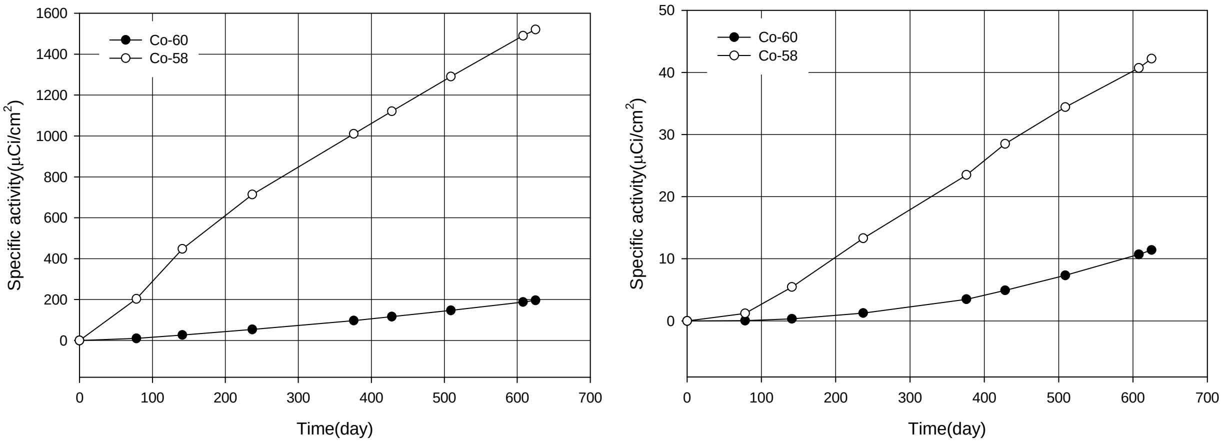


Fig. 3 Specific activities according to time (day) in the coolant in the core of the integral reactor.



(a) **(b)**

Fig. 4 Specific activities according to time(day) on the surface of (a) core and (b) steam generator in PLANT-I.

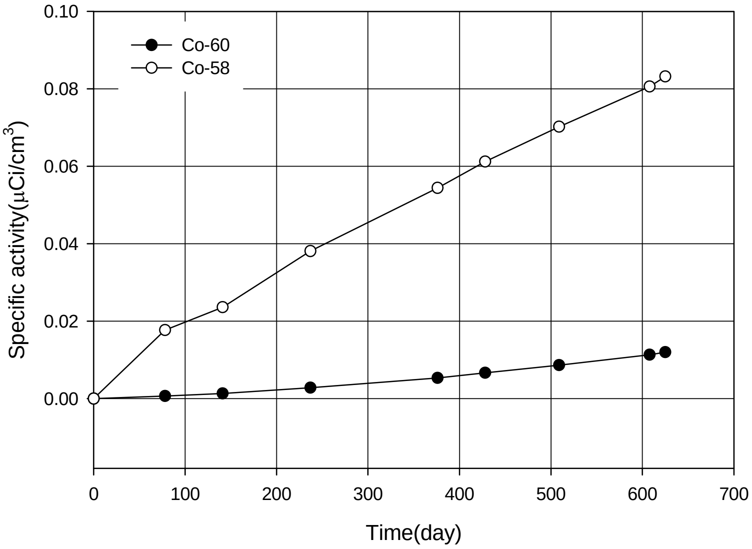
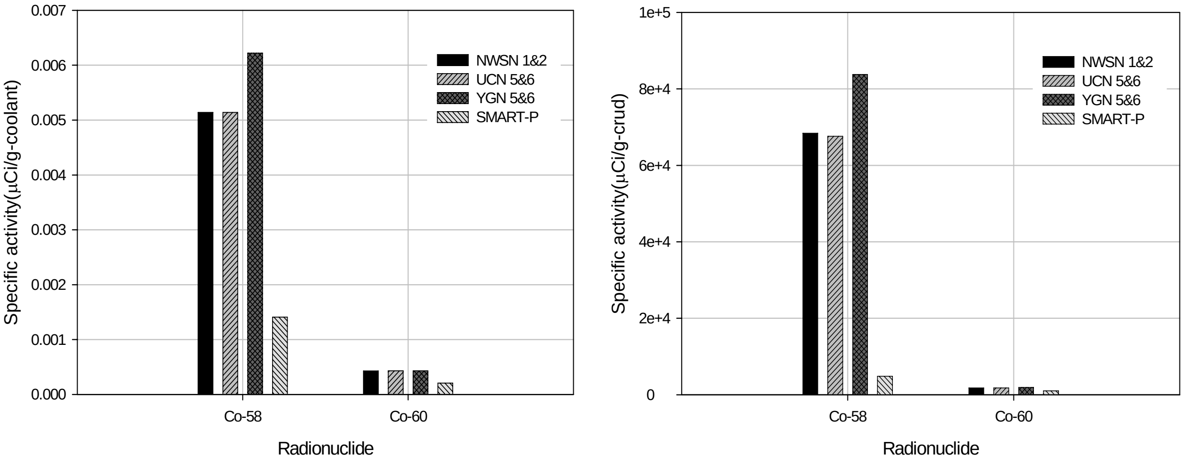


Fig. 5 Specific activities of Co-58 and Co-60 according to time (day) on the coolant in PLANT-I.

To compare with commercial nuclear power plants, specific activities are used in the New Wolsong Units 1&2, Yonggwang Units 5&6 and Ulchin Units 5&6. The specific activities of these power plants are given in their final safety analyst reports (FSAR), respectively. The specific activities of the Co-58 in the coolant of New Wolsong Units 1&2, Ulchin Units 5&6 and Yonggwang Units 5&6 are about 6.00×10^{-3} µCi/g as shown in **Fig. 6 (a)**. It was estimated as 1.41×10^{-3} µCi/g for the integral reactor, which is 1/4 times less than that of a KSNP. In the case of Co-60, the estimated specific activity of the integral reactor is similar to those of KSNP, but that of the integral reactor is the lowest as 2.06×10^{-4} µCi/g. The specific activities of Co-58 and Co-60 in surface are shown as **Fig. 6 (b)**. In case of Co-58, the specific activities of KSNP are about 8.00×10^{4} µCi/g, which is 1/10 times less than that of the integral reactor. The specific activity of Co-60 in the integral reactor is also the lowest. From this result we found that the design advantages of the water chemistry and steam generator material of the integral reactor can prevent and reduce the corrosion products in the primary circuit.

Finally, the relative contribution of the Co-58 and Co-60 nuclides to the specific activity varies according to plant operating time as shown in **Fig. 7**. Co-58, which has a half-life of 71 days, is the major contributor in the early stage of a plant operation. But after the early stage of the fuel cycle, this domination is reduced and gradually governed by the longer lived (5.26 year half-life) Co-60 isotope. In general, the long-term activity from the primary circuit in a plant attributes mainly to the accumulation of the Co-60 nuclides.



(a) **(b)**

Fig. 6 Specific activities between the integral reactor and the operating plants: (a) in primary coolant, (b) on the surface

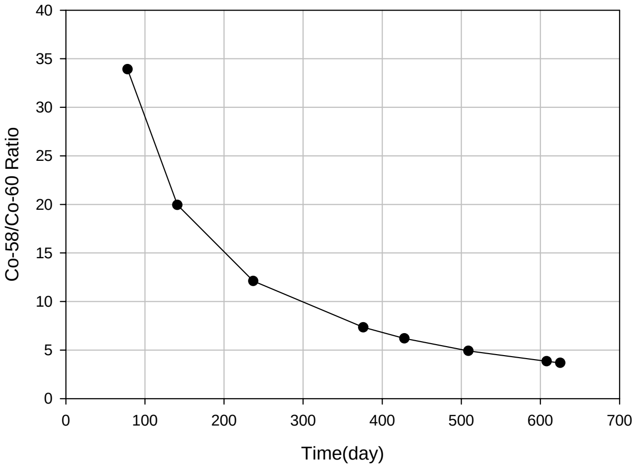


Fig. 7 Variation of Co-58/Co-60 ratio based on steam generator tube measurements in the integral reactor.

4. CONCLUSIONS & FURTHER STUDY

In order to evaluate the behavior of corrosion products in an integral reactor, a multi-region mass balance model is applied. The specific activities of the various species on the surface of the core and steam generator throughout the effective full power hours, 15,000 hours, are simulated for the integral reactor as well as PLANT-I, which has the same configuration as the integral reactor except that the water chemistry and steam generator material is the same as the KSNP. On the reactor core surface, the specific activity of Co-58 is estimated to be 26.5 µCi/cm^2. This is 30 times greater than that of the steam generator. Co-60 specific activity on the reactor core surface is 3.41 µCi/cm^2 , which is 16 times that of the steam generator. However, in the case of PLANT-I, it is expected to be very high compared to those of the integral reactor. It is considered that the higher values of the specific activities of Co-58 and Co-60 in PLANT-I are due to the higher release rate of Inconel-600 and the H_3BO_3-LiOH water chemistry condition. The titanium alloy for the steam generator tube material of the integral reactor has a high corrosion resistance compared to that of inconel. This is very helpful in reducing the coolant specific activity. Primary coolant standards for the KSNP provide for dissolved oxygen concentration conditions controlled by additives of hydrogen, if necessary. However, in the integral reactor, the hydrogen concentration is maintained by ammonia, which is used as a pH-reagent and dosed into the primary coolant where hydrogen and nitrogen are generated by a subsequent ammonia radiolytical and thermal decomposition.

As a result, the specific activities of the corrosion products in the integral reactor are very low compared to those of the KSNP such as the New Wolsong Units 1&2, Yonggwang Units 5&6 and Ulchin Units 5&6. In the integral reactor, a significant reduction of the specific activities from the activated corrosion products is obtained due to the introduction of the titanium alloy to the SG tube and the avoidance of a boric acid induced corrosion. From this result we conclude that the design features of the integral reactor can prevent and reduce the generation of corrosion products in the primary circuit.

Another purpose of this study was to establish a computational analysis system for the corrosion products behavior within a primary circuit for an integral reactor, which has an ammoniated water chemistry and the titanium alloy for SG tube material. From this point of view it is concluded that this program can be used as a engineering tool for analysis of the corrosion products within the primary coolant for the integral type reactor.

As mentioned earlier, the release rate of crud from structural materials has a lot of uncertainty. Even if a deposition rate coefficient is assumed from the experimental data, the value still has some uncertainty because the actual plant structure is very complicated. Thus, experiment for the measurement of the corrosion release rate should be continued to obtain the more precise data. As a further study, this program will be updated to simulate water chemistry effect and magnetite solubility as a function of the coolant pH.

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