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A study on Prediction of Radioactive Source-term from the Decommissioning of Domestic NPPs by using CRUDTRAN Code

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1. Introduction

It has been emphasized the importance of developing source term assessment technology of decommissioning and decommissioning owing to the permanent shutdown of the Kori #1, the life extension of the Wolsong #1, and the increase of deteriorated nuclear plants. This study sought the characteristics of materials of the primary system which has been the source of corrosion products based on the type of domestic NPPs. Among the nuclear plants, the study selected the Kori #1, a Westinghouse type nuclear plant to be decommissioned.

For the study, the behavior mechanism of corrosion products in the primary system of the Kori #1 was analyzed, and the volume of activated corrosion products in the primary system was assessed based on domestic plant data with the CRUDTRAN code used to predict the volume. It is expected that the study would be utilized in predicting radiation exposure of workers performing maintenance and repairs in high radiation areas and in selecting the process of decontaminations and decommissioning in the primary system. It is also expected that in the future it would be used as the baseline data to estimate the volume of radioactive wastes when decommissioning a nuclear plant in the future, which would be an important criterion in setting the level of radioactive wastes used to compute the quantity of radioactive wastes

2. Characteristics of Commercial NPPs in Domestic

2.1 Westinghouse

In Korea, there are several Westinghouse type power plants are currently in operation (such as the Kori #1, 2, 3, and 4; the Yeonggwang #1 and 2; and the Uljin #1 and 2) including the Kori #1. The Kori #1, the first nuclear plant in Korea, is decided to shut down permanently in June 2015. For materials used to make heat transfer pipes of steam generators, a major source of corrosion products in Westinghouse type nuclear plants currently in operation in Korea, Alloy-600 was used in the Kori #2, 3 and 4; the Yeonggwang #1 and 2; and the Uljin #1 and 2, while the steam generator of the Kori #1 was replaced in 1998 for the life extension. Before the replacement, the material of heat transfer pipe of the steam generator was Alloy-600, which was replaced with Alloy-690. Chemical composition of Alloy-600 (wt%) is as follows: more than 72 % of Ni, 14 ~ 17 % of Cr, 6 ~ 10 % of Fe, and under 0.15 % of C. Accordingly, corrosion reactions in the steam generator generate corrosion products such as Ni, Cr, and Fe, and then produce radioactive materials such as ^{58}Co, ^{60}Co, ^{54}Mn, ^{52}Cr, and ^{59}Fe owing to high temperature and radio-activation reactions as shown in Table 1. In Korea, materials used to make primary coolant pipes in every Westinghouse type power plant including the Kori #1 are wrought or casted by austenite stainless steel, SA 351.

2.2 CANDU

There are four 679-MW CANDU type pressurized heavy water reactors (PHWRs) in operation in Korea. PHWRs operating in Korea have around 380 nuclear fuel channels, and each channel forms a pressure boundary independently. These reactors have separate circulation systems: the primary coolant circulation system and the moderator circulation system. One of the characteristics is that there are pressures tubes filled with nuclear fuel bundles laid horizontally in calandria vessels made of Zr-2.5% Nb alloy, which are corresponding to nuclear reactor vessels in PWRs. A material used to make heat transfer pipes of steam generators which are main source of corrosion products in CANDU type nuclear plants currently in operation in Korea is Alloy-800. Unlike Alloy-600 and Alloy-690, both of which are Ni alloys, Alloy-800 is a high alloy steel containing a lot of Ni and Cr. Its chemical composition (wt %) includes 32.5 ~ 35.0 % of Ni, 21.0 ~ 23.0 % of Cr, and over 39.5 % of Fe. Accordingly, corrosion reactions in a steam generator produce corrosion products such as Ni, Cr, and Fe, which are converted into radioactive materials such as ^{58}Co, ^{60}Co, ^{54}Mn, ^{51}Cr, and ^{59}Fe owing to high temperature and neutron activation. The feeder pipe, another source of corrosion products in a CANDU type nuclear plant, is a main component of heat transport system of a CANDU type nuclear plant. It is used to transport pressurized heavy water through nuclear fuel channel (pressure tube) to remove heat generated by nuclear fission. Therefore, the material for feeder pipes are low carbon steel (SA 106 Gr.B) with corrosion resistance at high temperature, which contains under 0.3wt% of C, 0.29 ~ 1.06 % of Mn, and under 0.4 % of Cr. Accordingly, the flow accelerated corrosion reactions occurred in a feeder pipe generate corrosion products such as Fe, Mn, and Cr, and these corrosion products are converted to radioactive materials such as ^{54}Mn and ^{51}Cr owing to radio-activation reactions presented in Table 1. And various materials including zirconium alloy, carbon steel, stainless steel, and Ni alloy are used to make primary coolant pipes.

Table 1. Corrosion Product and generation source in NPPs

Nuclide	Half-life	Generation unit	Source
^{51}Cr	27.8d	$^{51}Cr(n,\gamma)^{51}Cr$	System material
^{54}Mn	312d	$^{54}Fe(n,p)^{54}Mn$	System material
^{59}Fe	45d	$^{59}Fe(n,\gamma)^{59}Fe$	System material
^{58}Co	71d	$^{58}Ni(n,p)^{58}Co$	S/G tube
^{60}Co	5.24y	$^{59}Co(n,\gamma)^{60}Co$	Stellite and Nickel Alloy

3. Corrosion Products

3.1 Generation of Corrosion Products

In NPPs, water is being used for diverse purposes including moderator of neutron, coolant for nuclear reactors and auxiliary feed water. The water is very chemically activated medium in a high temperature environment such as a nuclear power plant. From the experience of operating NPPs, it has been found that there are many phenomena occurred in normal operating conditions owing to water such as corrosion, erosion and depositions on system surfaces. The continual contacts between water and corroded surfaces in the primary system would mix corrosion products with coolant, which would be circulated in the system. Some of these corrosion products would be existed as a dissolved form, while some materials would become suspended solids made of insoluble metallic oxides to be deposited on the surfaces of pipes and equipment. It is called as CRUD, and the incidence rate of corrosions in NPPs is determined by temperature, systems, materials, environment of radiation field, durable years of materials, and so forth. In general, it is known that in the initial stage of operating a NPP, the corrosion incidence rate is increasing steeply and over time it would be in a constant equilibrium state. Since the coolant system contains fuel, it is the major source of radioactive materials. In a coolant system, any metal contacting with the coolant becomes corroded slowly. Some corrosion products may be dissolved or suspended in the coolant, or passed through neutron fluxes in the reactor core. As corrosion products between coolant and system surfaces have been exchanged continuously, the radioactive materials have been accumulated progressively on the system surfaces ultimately, and radiation fields have been formed. Some radioactive materials have leaked from the coolant system and have become the secondary source of radiation.

3.2 Deposition of Corrosion Products

Although the CRUD concentration in primary coolant is rather low, since the coolant is flowing at high speed, there may be a considerable quantity of CRUD may be moving into the core in a relatively short period of time. And the CRUD is unstable at all times, it tends to be deposited on any part of the coolant system while moving. Such CRUD has the following characteristics:

- Deposited at parts with high heat flux
- Increased deposition of CRUD in radiation flux
- Increased deposition of CRUD at parts with low fluid velocity
- Heavily deposited on the surface of Zircaloy rather than that of stainless steel
- Less deposition in operating conditions of high pH than in neutral pH

The quantity of CRUD generated is increasing following the operations of NPPs. If it is deposited on nuclear fuel cladding, it may curtail the capacity of heat transfer, and as it contains a considerable quantity of boron, it may cause an unbalance of output. If the CRUD is deposited in tubes of a steam generator, it will reduce the capacity of heat transfer, and increase the radiation level in the steam generator, which would make its maintenance very difficult. In addition, if CRUD is deposited in some low fluid velocity parts of mechanical components such as control rod actuators or valves, then it may cause mechanical damage

3.3 Radio-Activation of Corrosion Products

A considerable quantity of CRUD in the coolant of a primary system would be deposited on the reactor core surface through continuous circulation. When such materials are activated, it will be converted into a material with intense radioactivity. Through sequential substitution of these materials with CRUD in the coolant continuously, the CRUD becomes radioactive intensely. Such generation of radioactive corrosion products is affected by factors such as the neutron flux, the number of atoms of the target isotope, the cross section for the reaction and the time of irradiation. Radioactive nuclides, ^{58}Co and ^{60}Co, which exert the largest influence over the radiation exposure of workers among the corrosion products, are generated by the irradiation of thermal neutrons on ^{58}Ni and ^{59}Co. Especially, ^{60}Co has very long half-life (around 5.3 years) and emits high energy gamma radiation. ^{59}Co exists as an impurity of Ni, a component of Inconel and stainless steel, or contains in stellite as its basic component. There have been many researches on the fundamental reduction of ^{59}Co contained in metallic materials of the primary system, as a method of reducing the level of radiation in a NPP. It has been recognized as an effective method of decreasing radiation sources along with the water quality control.

4. Behavior Mechanism of Corrosion Products in PWR

4.1 PWR

The primary coolant system of the PWR can be largely divided into Core, Coolant and S/G. The major driving force of behavior of the corrosion products in the primary coolant system is the change of solubility according to the change of coolant temperature. Figure 1 shows behaviors of corrosion products and radioactive materials in the primary coolant system. CRUD type metallic impurities are discharged into the condensate water or feed water from the surfaces of parts or pipes in the system. These impurities would be circulated and entered into the reactor coolant system.

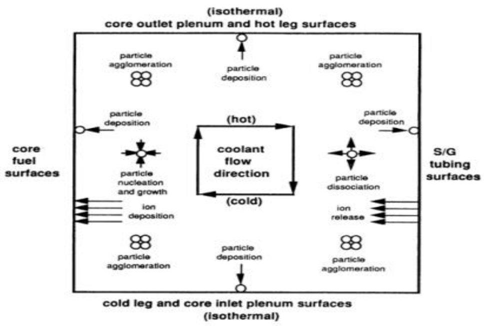

Fig 1. Process involved in PWR Corrosion Products Transport

Metallic ions are discharged into the coolant from corroded surfaces in the primary system, and those which exceed the saturation level of the coolant would form particle materials such as colloids or oxides or non-oxides. Owing to the interactions between soluble ions and CRUD particles, ionic materials are adsorbed on the surfaces of CRUD particles suspended in the coolant. These CRUD and soluble materials become precipitated on fuel surface by diverse mechanisms: some of these materials are not leaked in the system and fixed over the fuel surface and radio-activated by neutron irradiation from precipitates on the fuel surfaces and structure material in the reactor core. Owing to dissolution and friction, radioactive materials are discharged from structural material in the reactor core, while owing to erosion and exfoliation of precipitates on the fuel surface owing to water pressure, radioactive materials are dissolved into the coolant. The radioactive materials dissolved into the reactor coolant system become redistributed as dissolved materials and particulate materials based on the solubility of oxides and the concentration of metallic corrosion products acting as collectors. These radioactive materials become precipitated on outer surface of the reactor core owing to various interactions. There are two layers on the outer shell of the reactor core formed by the corrosion of base metal and the CRUD precipitates containing water. Owing to oxidation on the surface, some corrosion products are leaked into the coolant from the oxidized layer on the outer surface of the reactor core.

5. Application to Domestic NPPs

5.1 Selection of Code and Its Background

The CRUDTRAN code was developed by the Massachusetts Institute of Technology (MIT) and has been used to analyze and predict corrosion products in the primary coolant system of a PWR and behaviors of radioactive nuclides such as ^{58}Co and ^{60}Co. The CRUDTRAN code could predict the movements of corrosion products based on the difference in solubility, locomotive force and other experimental variables. It could interpret the impacts caused by chemical changes of the coolant well when the corrosion products are moving. The CRUDTRAN code divides a nuclear plant into three major zones: reactor core, coolant, and steam generator. This study would contribute to researches on prediction of decommissioning source term conducted in the future by comparing the modeling results on a PWR, PHWR (the Kori #1, Wolsong #1) with the CRUDTRAN code and the data actually measured at the Kori #1 and check the similarity to get the level of confidence

5.2 Application to PWR

5.2.1 Selection of Cycle

As mentioned above, this study selected a specific cycle based on data of water chemistry and the primary system of the Kori #1 among NPPs in domestic, modelled by using the CRUDTRAN code, and then compared and analyzed the behavior of the corrosion products and radioactive materials generated in the primary system. In here, the specific cycle is from the date of starting commercial operations of the Kori #1 (1978) through the entire life cycle and the total inventory of radioactive materials over time has been assessed. To verify the similarity by using the CRUDTRAN code, the level of confidence was assessed by comparing and verifying the average measured values of ^{58}Co and ^{60}Co during the operational duration (between 2006 and 2015) presented in the in-house data of the Korea Hydro & Nuclear Power Co., Ltd. (KHNP) and the average estimated values computed with the CRUDTRAN code. The number of factors required for the CRUDTRAN code is around 50 in total, which were collected from various reports including actual measurement data of the Kori #1 offered by the KHNP.

5.2.2 Assessment Results

Figures 2 show the results of analysis of radioactive nuclide inventory in each zone during the entire cycle of the Kori #1, including the behavior changes of ^{58}Co and

^{60}Co in the reactor core, the steam generator, and the coolant. The modeling results show that the inventory of radioactive nuclides has been fluctuating periodically according to the cycle of preventive maintenance period. Figure 3 is a graph showing the similarity obtained by comparing and verifying the average estimated values computed by using the CRUDTRAN code with the average measured values of ^{58}Co and ^{60}Co in the primary system by year as presented in the in-house data of the KHNP (measured between 2006 and 2015). The modeling shows that when comparing the measured values and the estimated values, they are encompassed in the range of allowable errors and revealing a similar trend.

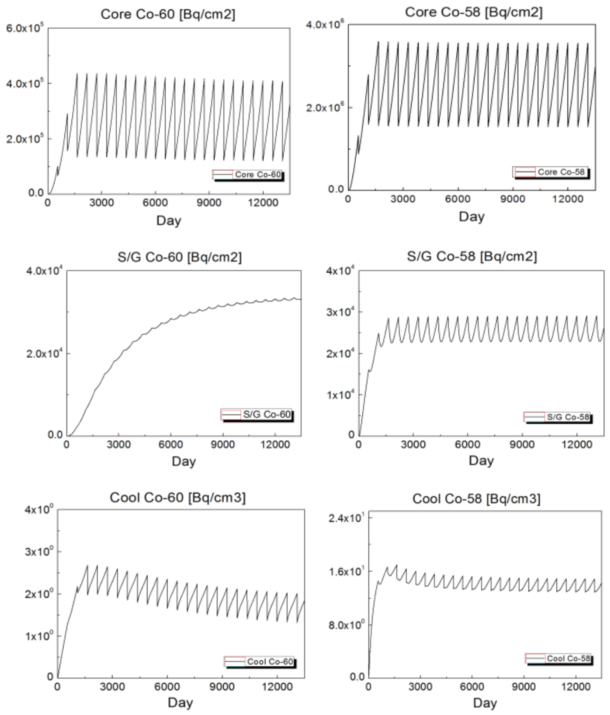

Fig 2. The variation of the Co concentration in the primary system during life-cycle (Core, S/G, Coolant)

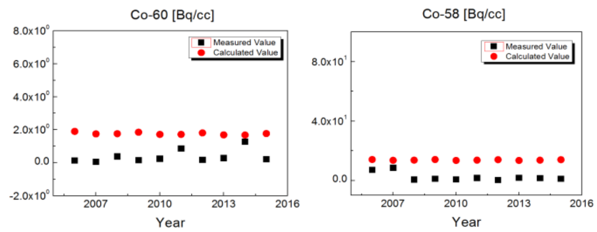

Fig 3. The comparison of measured and calculated values at Kore #1

6. Conclusions

This study analyzed the material properties of the primary system equipment, which are sources of corrosion products following to the type of domestic NPPs (such as Westinghouse, CANDU) as basic data for the assessment of radiation source term from the decommissioning of NPPs. In the study, a Westinghouse type NPP decided to decommission was selected.

Accordingly, properties and behavior mechanisms of radioactive corrosion products in the Westinghouse type NPP were analyzed. It was found out that the major driving force of behavior of corrosion products in the primary coolant system of the PWR is the change of solubility owing to the temperature change of the primary coolant. To predict and assess the radioactive nuclide inventory in the primary system, the CRUDTRAN code was applied. From the interpretation of the results, a fluctuation trend of radiation source term caused by the operating cycle was confirmed, and the level of confidence of the code was verified as both the on-site measured values and the estimated value from the code were determined to be within the error range, although there were some differences.

The results of prediction of the radioactive nuclide inventory in the primary system performed in this study would be used as baseline data for the estimation of the volume of radioactive wastes when decommissioning NPPs in the future. It is also expected that the data would be important criteria used to classify the level of radioactive wastes to calculate the volume. In addition, it is expected that the data would be utilized in reducing radiation exposure of workers in charge of system maintenance and repairing in high radiation zones and also predicting the selection of decontaminations and decommissioning processes in the primary systems. In future researches, it is planned to conduct the source term assessment against other NPP types such as CANDU and OPR-1000, in addition to the Westinghouse type nuclear plants. But there were many difficulties in acquiring on-site data of the relevant NPPs when assessing by using the code. Therefore, if it is possible to acquire on-site data as much as possible in the future, then it will be possible to conduct a more reliable study on the assessment of decommissioning source term.

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