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Study on the deriving considerations for selecting system decontamination methods for nuclear power plant decommissioning

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1. Introduction

Radioactive waste generated in the primary system of nuclear power plants is radioactivated by neutrons during operation, and the management of these materials is very important in terms of radioactive waste management. In particular, the management of radioactive corrosion products caused by neutrons on the surface of nuclear fuel and inside the primary system is very important in terms of ensuring the soundness of equipment during operation, water chemistry management, and worker exposure management during operation.

Corrosion products are present as radioactive corrosion products due to neutron irradiation from the surface of the nuclear fuel in the core, and the nature of the radioactivity due to neutron irradiation varies depending on the components of the structure. Therefore, it is very important to obtain accurate information on the source and its characteristics.

The main components of radioactive corrosion products are compounds containing Fe, Cr, and Ni as the main species, with Co as the main component of the crud. In general, stainless steel and Inconel materials are reported to contain around 0.1% Co.

In this study, we derived compound forms that can be considered as a preliminary study for the selection of a systematic decontamination method for the removal of radioactive corrosion products present in surface contamination.

2. Methods and Results

2.1 Survey/Analysis of System Decontamination Methods

Representative chemical decontamination methods for removing corrosion oxides generated on metal surfaces include chelates and organic acids, strong inorganic acids and chemical agents, oxidizing agents and reducing agents, and TechXtract, and the method of cross-application of oxidizing and reducing agents is mainly effective. In particular, reductive methods for low-concentration chemical decontamination are categorized into the CAN-DECON process, which uses a complexing agent (organic acid, EDTA, etc.), and the LOMI process, which uses an acid solution as a complexing agent with metal ions as a reducing agent. It has been investigated that the dissolution reaction of metal ions in the oxidizing salt process is Cr, Ni, and Fe in the order of Fe, Cr, and Ni in the reducing salt process, and the dissolution reaction is close to equilibrium.

As an application example, developed countries with experience in decommissioning nuclear power plants have developed DfD, HP/CORD UV, LOMI, CANDEREM™, CITROX, REMCON, NITROX, etc. for system decontamination, and the principle of decontamination considering the compound form according to the step-by-step chemical decontamination process is shown in Figure 1. Field application experience in decommissioned nuclear power plants includes the use of DfD technology as a controlled decontamination technology at Maine Yankee and Big Rock Point nuclear power plants, CORD technology at Connecticut Yankee nuclear power plant, and mixed method (NITROX+DfD) at Jose Cabrera nuclear power plant.

2.2 Selection of Evaluation Codes

In this study, considering the situation in which experiments to derive the form of compounds existing in the crud in the primary system could not be performed, we attempted to use evaluation codes to identify the forms of compounds that could exist.

Thermodynamic codes based on thermochemical databases such as HSC Chemistry, FactSage, Thermo-Calc, MULTEQ V9, etc. were investigated to derive the compound forms of corrosion products generated in nuclear power plants, and the TPP module in HSC Chemistry was selected for this study.

The TPP Diagram module, which is used in this paper, is a diagram that shows the stable phase under isothermal conditions consisting of three elements based on Gibbs energy as the partial pressure of two components and calculates the phase stability diagram using partial pressure on two axes or temperature on the x-axis and partial pressure on the y-axis using the Gibbs energy minimization method.

2.3 Deriving Input Factors

2.3.1 Selection of primary system material

Alloy alloy steel, which is the material of the main components of the primary system, was selected as the standard for evaluation, and the main component information was referenced to the Mill Test Certificate.

Among the main constituent elements, compound forms were derived, including those that contribute to radioactivity and those that are considered as decelerants and additives in nuclear power plant operations. In addition, since hard water (H_2O) is used in the cooling system, we considered the environment containing hydrogen (H) and oxygen (O) in addition to the metallic components.

2.3.2 Establishment of radioactive corrosion product derivation evaluation environment

Various environments where elements of Cr, Zn, Fe, Ni, Co, and Li, B, H, and O, which are used as additives and coolants in the operation of nuclear power plants, and elements that can be generated as radioactive corrosion products, were selected as evaluation targets and input factors.

2.4 Evaluation Results

The conformations of the compounds derived using the evaluation code are shown in Table 1, and the main conformations of the compounds were confirmed to exist by surveying the literature.

Table. 1. Results of compound conformation in radioactive corrosion products

Compound Forms
Ni_2FeBO_5,$LiBO_2$, H_3BO_3, $Fe_2Fe(BO_3)O_2$, Fe_3BO_6, $Li_2B_2O_4$, $Ni_2Fe(BO_3)O_2$, $Li_2NiFe_2O_4$, $LiFeO_2$, $LiBO_2$, $ZnFeO_4$, $NiCr_2O_4$, $FeCr_2O_4$, $ZnCr_2O_4$, $CoFe_2O_4$, FeB_4O_7, $Fe_2B_2O_5$, Fe_3BO_5, CoB, Co_2B, $LiCrO_2$, Li_2CrO_4, Li_3CrO_4, $LiFeO_2$, $LiFe_5O_8$, $Li_2Fe_3O_5$, Li_5FeO_4, $Li_2O*5Fe_2O_3$, $LiCoO_2$, $ZnCrO_4$, $ZnCr_2O_4$, ZnO, $ZnO*Cr_2O_3$, $Fe_{0.08}Zn_{0.92}$, $Fe_{0.12}Zn_{0.88}$, $Fe_{0.21}Zn_{0.79}$, $Fe_{0.31}Zn_{0.69}$, $Zn_{0.1}Fe_{2.9}O_4$, $Zn_{0.3}Fe_{2.7}O_4$, $Zn_{0.5}Fe_{2.5}O_4$, $Zn_{0.7}Fe_{2.3}O_4$, $ZnFe_2O_4$, $ZnFe_{29}O_{40}$, $Zn_3Fe_{27}O_{40}$, $Zn_5Fe_{25}O_{40}$, $Zn_7Fe_{23}O_{40}$, $ZnCo_3$, FeO, Fe_2O_3, Fe_3O_4, NiO, CrO_2, CrO_3, Cr_2O_3, Cr_3O_4, Co_3O_4, $NiFe_2O_4$, $Cr_{22}Ni_8$, Cr_5O_{12}, Cr_8O_{21}, $NiO*Cr_2O_3$, Cr_2FeO_4, Fe_8Cr_{22}, $Fe_{26}Cr_4$, $FeCr_2O_4$, $CoCr_2O_4$, $CoO*Cr_2O_3$

3. Conclusions

In this study, an evaluation code was utilized to derive the morphology of the compounds present in the radioactive corrosion products to suggest the need for selecting the optimal decontamination technology.

The results obtained are the basis for evaluating the compound behavior of nuclides that may be present in the crud, but since the results are programmatic, it is necessary to compare the results with the results of the evaluation reflecting the analytical data of the crud generated during future decommissioning. These results may be meaningful in reducing potential risks in the selection of technologies for system decontamination.

However, the compound containing boron in the crud is bonaccordite, and the increase in deposition of the target compound was found to be over 400 ° C for a thickness of 59 µm. Since the coolant temperature typically varies between 280 and 320 ° C during reactor operation, boron-containing compounds can be excluded. Further studies should consider the morphology of the compounds identified in this paper when selecting a system decontamination technology for future decommissioning.

The results derived in this paper are basic data evaluating the compound behavior of nuclides that may exist in crud. Since the evaluation results are the result of using a program, it is necessary to compare them with the evaluation results reflecting the analysis data of crud generated during future decommissioning. do. These results are expected to be meaningful in reducing potential risks in technology selection for future system decontamination.

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