

Analysis of Characteristics of Spent Fuels on Long-Term Dry Storage Condition

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Currently, the interim storage pools of spent fuels in South Korea are expected to become saturated from 2024. It is required to prepare an operation plan of a domestic dry storage facility during a long-term period, with the researches on safety evaluation methods. This study modified the FRAPCON code to predict the spent fuel integrity evaluation such as the axial cladding temperature, the hoop stress and hydrogen distribution in dry storage. The cladding temperature in dry storage was calculated using the COBRA-SFS code with the burnup information which was calculated using the FRAPCON code. The hoop stress was calculated using the ideal gas equation with spent fuel information such as rod internal pressure. Numerical analysis method was used to calculate the degree of hydrogen diffusion according to the hydrogen concentration and temperature distribution during a dry storage period. Before 50 years of dry storage, the cladding temperature and hoop stress decreased rapidly. However, after 50 years, they decreased gradually and the cladding temperature was below 400 K. The initial temperature distribution and hydrogen concentration showed a parabolic line, but hydrogen was transferred by the hydrogen concentration and temperature gradient over time.

Keywords: Dry storage, FRAPCON code, Cladding temperature, Hoop stress, Hydrogen distribution

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1. Introduction

Spent fuels discharged from pressurized water reactors around the world have been stored in various dry storage facilities during long-term periods. Final disposal plans for spent fuels have been established in only a few countries. The period of dry storage in the United States of America has been extended from 20 years to 40 years because of the closure of final disposal project in the Yucca Mountain high-level radioactive waste repository. Currently, the interim storage pools of spent fuels in South Korea are expected to become saturated from 2024 [1]. However, it has been delayed to obtain high consent of the public on the publicizing process of spent fuels. Moreover, there is no experience in operating any dry storage facility for storage of spent fuels discharged from domestic pressurized water reactors. Thus, many preparations for dry storage are required in various aspects. In addition, there is no final disposal plan to be implemented after dry storage of spent fuels. Thus, it is also required to prepare an operation plan of a domestic dry storage facility during a long-term period, with the research on safety evaluation methods. In particular, as the change of the characteristics of spent fuel depends on a period of dry storage, it is critical to make accurate predictions of fuel cladding temperature, hoop stress change, and hydrogen behavior and assess the integrity of spent fuels. Spent fuels have various characteristics depending on their irradiation histories. Thus, this study developed the modified FRAPCON-4.0 code to simulate the axial characteristics (temperature, hoop stress, hydrogen distribution) of spent fuels during a dry storage period, and analyze the behavioral changes of spent fuels.

The Nuclear Regulatory Commission in the United States limits the maximum temperature of spent fuel with burnups exceeding 45 GWD/MTU to 400°C in dry storage condition. The purpose of the restriction is to prevent damages in the spent fuel cladding and the components of dry storage system due to this temperature. Cladding damage occurring at this temperature is highly associated with

brittleness caused by radial hydride reorientation. Hydrogen brittleness occurs in a high-stress and high-temperature condition, and sensitizes the cladding to grow cracks during a long-term dry storage. The temperature limit of 400°C, which was determined based on many researches to prevent hydride reorientation, has been implied to limit the hoop stress under 90 MPa. In other words, it is not necessary to analyze the hoop stress as long as the temperature is maintained below 400°C. Accordingly, all manufacturers of dry storage casks have kept the temperature below 400°C. Nevertheless, the ISG-11 document shows that it is important to analyze the hoop stress at 400°C to ensure the hoop stress of a high-burnup fuel cladding maintain below 90 MPa [2-5].

The purpose of this study is to identify an analysis method for the characteristics of spent fuels on a long-term dry storage condition and to analyze their characteristics according to a period of dry storage using the FRAPCON code that can simulate the characteristics of spent fuels in dry storage. Using the FRAPCON-4.0 code that allows users to modify its FORTRAN sources, the characteristics behaviors of spent fuels on a long-term dry storage condition were analyzed. The axial temperature, hoop stress, and hydrogen concentration of the spent fuel cladding in a long-term dry storage condition were predicted, and a program for the calculation processes were developed.

2. Analysis Method

2.1 Key factors of spent fuel characteristics in dry storage condition

2.1.1 Hoop stress

The hoop stress of a spent fuel cladding is a stress exerted circumferentially in a cylinder wall that is under internal pressure. In a spent fuel cladding, the hoop stress keeps changing according to change in internal pressure due to fission gas release and change in diameter due to oxidation. Thus, the hoop stress is regarded as a key factor in evaluating

the integrity of a spent fuel cladding. The hoop stress (σ) of a spent fuel cladding is defined as follows:

$$\sigma = \frac{r_i P_i - r_o P_o}{t}, \quad (1)$$

where r_i and r_o are the inner and outer radius of a spent fuel cladding, respectively. P_i and P_o are the inner and outer pressure of a spent fuel cladding, respectively. t is the thickness of a spent fuel cladding [5-7].

The main factor of the hoop stress is the internal pressure of a spent fuel rod. The internal pressure generated by fission gases released during nuclear reactor operation has a strong effect on cladding degradation. Because it is a direct factor triggering cladding creep, hydride reorientation, and delayed hydride cracking on a long-term dry storage condition. In addition to the oxide layer thickness and hydrogen concentration of a spent fuel cladding, the internal pressure of a spent fuel rod depends on the amount of fission gas release according to the irradiation history of a spent fuel. Its internal pressure increases as burnup increases, and a high bias in the internal pressure could occur according to the irradiation history even if burnups are the same. Thus, the irradiation history as well as burnups is an important factor when the criteria to classify spent fuels is determined [5-7].

2.1.2 Change in cladding thickness

A fuel cladding undergoes an oxidation reaction in the coolant of a pressurized water reactor according to Eq. (2);



When an oxidation reaction occurs, an oxide layer and hydrogen are generated on zirconium of a fuel cladding [5-7]. The oxide layer serves as an insulator for nuclear fuel and increases the radius of the fuel cladding. The change ratio of zirconium thickness is 1.56 according to the Pilling-Bedworth ratio. This factor affects hoop stress of a fuel cladding. The effective thickness of a fuel cladding (t_{eff}) can be calculated as follows:

$$t_{eff} = t_{nom} - \frac{\delta}{1.56}, \quad (3)$$

where t_{nom} is the nominal thickness of a fuel cladding and δ is the thickness of an oxide layer [5].

The thickness of the oxide layer on a fuel cladding increases as the burnup increases. A high bias in the thickness of the oxide layer could occur when the burnup exceeds 40 GWd/MTU. Thus, the thickness of the oxide layer depends on an irradiation history even if burnups are the same [6-7].

2.1.3 Hydrogen generation

During the operation of a pressurized water reactor, zirconium alloys undergoes an oxidation reaction in coolant and then absorb hydrogen generated by this reaction. Hydrogen absorption is a phenomenon where hydrogen generated by an oxidation reaction is partly absorbed into zirconium of a fuel cladding. Hydrogen is produced by corrosion of zirconium oxidized in coolant. The equation about hydrogen generation represents in Eq. (2). The fraction of absorbed hydrogen is proportional to the oxidized hydrogen volume of the total hydrogen accumulated inside the cladding. Zircaloy-4, which is the fuel cladding of a pressurized water reactor, has a fraction of absorbed hydrogen between 0.15 and 0.16. The content of hydride increases as the burnup of a spent fuel increases [5-7].

2.2 Analysis method for behavior of spent fuel using FRAPCON code

2.2.1 Temperature prediction

The temperature distribution of a dry storage cask with spent fuel assemblies was calculated using the COBRA-SFS code. The COBRA-SFS code is widely using the temperature prediction of spent fuels since its feasibility was verified through the comparative experiment on the TN-24P storage cask. Considering the characteristics of a spent fuel depend on irradiation history, the temperature was calculated using the irradiation data of a specific spent fuel based

on the COBRA-SFS code [8-10].

The FRAPCON code was used to obtain the irradiation data. FRAPCON-4.0 code is the fuel performance code in reactor and also calculates the peak cladding temperature in dry storage through spent fuel module [11]. The irradiation data of the FRAPCON code included the average burnup, maximum burnup, and axial heat rating data. Among data, the most essential data for the COBRA-SFS code were the data on heat sources. Thus, the average burnup of the FRAPCON code was converted into the heat source data with time using the Eq. (4) for heat source by burnup and time, derived in the previous research through the ORIGEN code [12];

$$q = a \cdot t^{-b}, \quad (4)$$

$$a = 6.76 \times 10^{-2} \cdot Bu + 1.48, \quad (5)$$

$$b = 7 \times 10^{-5} \cdot Bu^2 - 9.4 \times 10^{-3} \cdot Bu + 0.97, \quad (6)$$

where q (kW) is the heat source per fuel assembly, t is time, and Bu is the average burnup of a spent fuel. For the data on dry storage casks and fuel assemblies required to the COBRA-SFS code, the TN-24P storage cask and the Westinghouse (WH) 15×15 fuel assembly were applied [10].

2.2.2 Hoop stress prediction

The axial hoop stress distribution at the final burnup of spent fuel obtained by the FRAPCON-4.0 code was applied to the initial hoop stress of a cladding during a storage period of spent fuel. The hoop stress during fuel irradiation was also calculated by the FRAPCON code using the internal pressure of a fuel rod due to fission gas release and cladding thickness change due to an oxidation reaction. Then, the hoop stress was calculated using the temperature change of dry storage cask according to the ideal gas equation.

2.2.3 Hydrogen concentration distribution prediction

The axial hydrogen concentration distribution at the final burnup of spent fuel obtained by the FRAPCON-4.0 code was applied to the initial hydrogen concentration in a cladding during a storage period of spent fuel. The hydrogen concentration during fuel irradiation was also calculated by the FRAPCON-4.0 code using the amount of hydrogen absorbed by an oxidation reaction of a fuel cladding and fuel temperature. Subsequently, the hydrogen concentration of irradiated fuel was predicted by applying the phenomenon where hydrogen is diffused by temperature and concentration gradient on a dry storage condition. The prediction was performed using the numerical analysis with the Fick's law related to diffusion by concentration difference and the Soret effect related to concentration gradient by temperature difference;

$$D = D_0 \cdot \exp\left(\frac{-Q}{RT}\right), \quad (7)$$

$$J = -D \nabla C - D \frac{QC}{RT^2} \nabla T, \quad (8)$$

where D is the diffusion coefficient of hydrogen in a fuel cladding and Q is the activation energy for hydrogen diffusion. R is the universal gas constant and T is temperature. J is the flux of diffusing atoms and C is the solid solution concentration of hydrogen in a fuel cladding. The diffusion of hydrogen in a fuel cladding mainly affects the diffusion coefficient of hydrogen, concentration gradient, and temperature gradient. The diffusion coefficient of hydrogen increases as temperature increases [13]. The numerical analysis was conducted using the Forward in Time Central Space (FTCS) scheme that reflects time and space. This method was derived from the Euler's Law [14].

2.3 Modification of FRAPCON code

The FRAPCON-4.0 code with the FORTRAN sources was modified to enable the calculation of the axial temperature, hoop stress, and hydrogen diffusion of a spent fuel cladding on a long-term dry storage condition. Fig. 1 shows

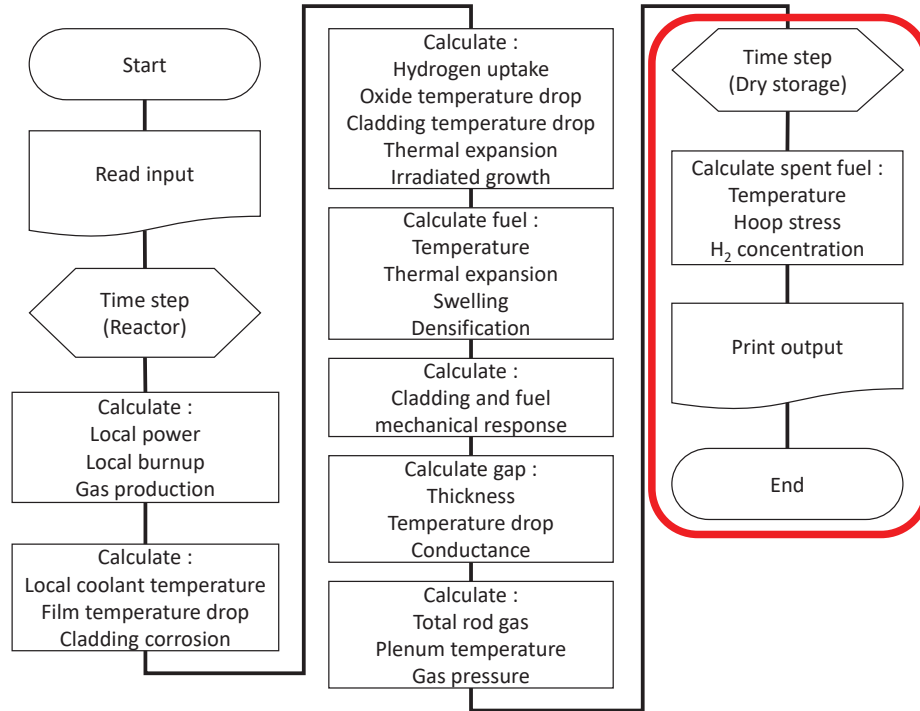


Fig. 1. Flow chart of modified FRAPCON-4.0 code.

the flow chart of the modified FRAPCON-4.0 code, where the modified parts are highlighted in red.

For the temperature prediction, the temperature prediction equation was drawn by the COBRA-SFS code using the data of a dry storage cask and fuel assemblies, and heat source and heating rating values of each spent fuel assembly. In general, the calculation time required by the COBRA-SFS code increases according to the number of axial nodes designated for calculation. For example, when 35 axial nodes are designated, approximately 30 minutes will be required to calculate the fuel temperature at one axial node. On the other hand, one of the advantages of the FRAPCON-4.0 code can simulate fuel irradiation in a short time. Thus, this study chose the method to derive a temperature prediction equation and apply it to the FRAPCON-4.0 code, rather than connecting the FRAPCON-4.0 code with the COBRA-SFS code and performing iterative calculations. As a result, the calculation time took approximately ten

seconds. To derive a temperature prediction equation, the FRAPCON-4.0 code required an average burnup and axial heating rating values. The average burnup was converted into the heat source value through the equation derived using the ORIGEN code. However, there was a problem of deriving the temperature prediction equation since even if many fuels have the same burnup, the heat rating values according to their irradiation histories could differ. To overcome this problem, the axial heat rating values of various fuels suggested in Ref. 15 were considered. Among them, the data of the spent fuel with a burnup of 46 GWD/MTU or higher were applied to perform the behavioral analysis of high-burnup spent fuels. In this way, it was possible to derive the temperature prediction equation of the spent fuel of 46 GWD/MTU or higher on a long-term dry storage condition as follows:

$$T = Y_0 + A_1 \times \sin \left\{ \pi \cdot \left(\frac{i - x_c}{w} \right) \right\}, \quad (9)$$

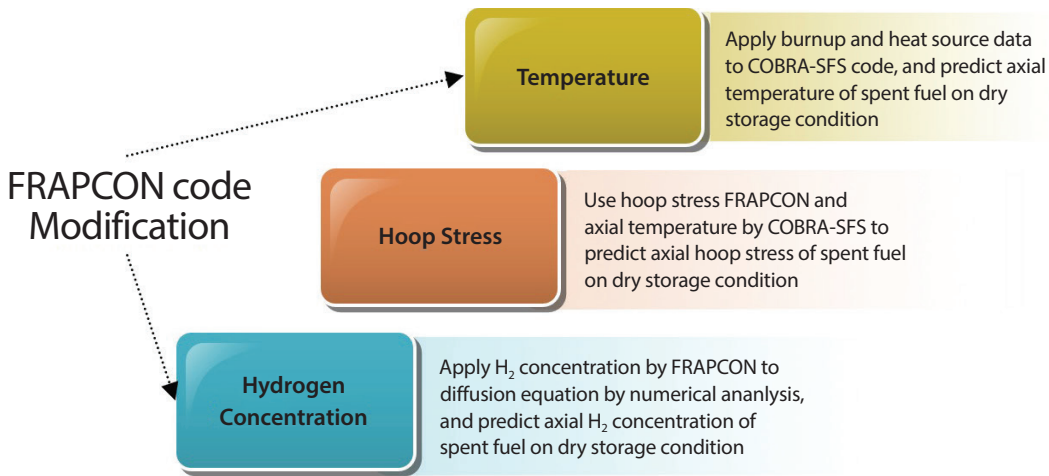


Fig. 2. Overview of modified FRAPCON code.

$$Y_0 = 4,728,650 \times \exp\left(\frac{-q}{2,008.38}\right) - 4,753,930, \quad (10)$$

$$A_1 = -4,729,900 \times \exp\left(\frac{-q}{2,008.49}\right) + 4,755,270, \quad (11)$$

$$x_c = 5.46 \times \exp\left(\frac{-q}{348.49}\right) - 61.55, \quad (12)$$

$$w = -10.96 \times \exp\left(\frac{-q}{345.06}\right) + 124.26, \quad (13)$$

where i is the ratio of the axial height of a spent fuel and q is the heat source of a spent fuel. The average error between the derived temperature prediction equation and the COBRA-SFS code was within 4.0%, and the peak cladding temperature was within approximately 3.0%.

For the hoop stress prediction, a pressure change in accordance with a temperature change of spent fuel cladding was calculated using the ideal gas law to evaluate the hoop stress on a dry storage condition. The hydrogen concentration prediction was performed applying the diffusion of hydrogen that was absorbed into the fuel cladding among the total hydrogen generated from the irradiated fuel. It was performed using the FTCS scheme that reflects time and space. This method is a numerical analysis based on the Euler's law. The average error rate during the dry storage

period of 300 years was within 1.5%. The method required to designate at least 35 to 45 positions. The number of axial regions along the fuel rod in the FRAPCON-4.0 code also had to be set to the same positions. In addition, excessive hydrogen concentration in the fuel cladding was applied to calculate the ratio of dissolved hydrogen considering temperature and hydrogen concentration with time. Fig. 2 shows the overview of the modified FRAPCON code.

3. Analysis Result

This study evaluated based on an irradiated fuel of 54.7 GWD/MTU with the enrichment of 3.2 w/o, a dry storage cask backfilled with helium in the cavity, and the cooling time of 10 years in a storage pool as shown in Table 1. Subsequently, the behaviors of the spent fuel for 300 years were analyzed [12].

3.1 Temperature prediction

The temperature distribution was predicted using the modified FRAPCON-4.0 code with the temperature prediction equation of Eq. (9). The temperature prediction

Table 1. Input parameters [10]

Parameter	Value	Parameter	Value
Cladding outer diameter	10.71 mm	Assembly type	WH 15×15
Cladding thickness	0.62 mm	Average burnup	54.7 GWd/MTU
Fuel U-235 enrichment	3.2 w/o	Cooling type	Helium
Cladding type	Zircaloy-4	Cooling time	10 years

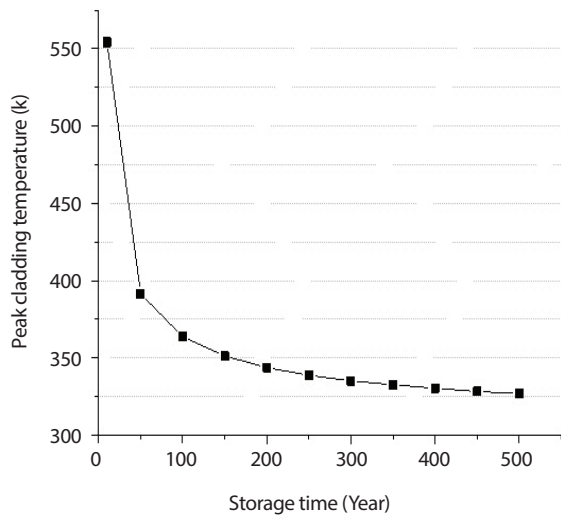


Fig. 3. Peak temperature of spent fuel cladding.

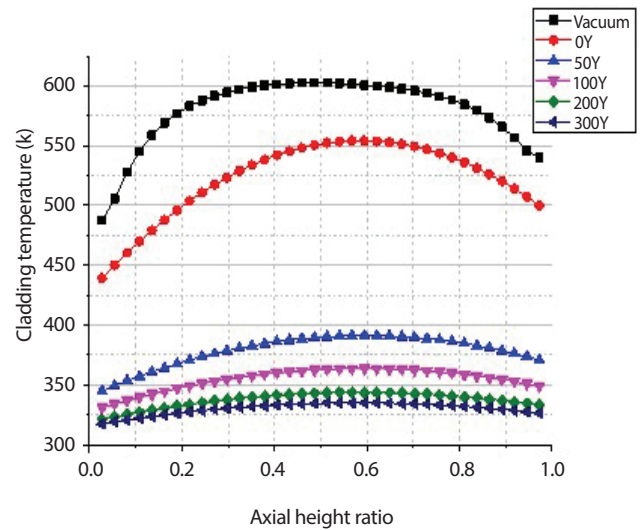


Fig. 4. Axial temperature distribution of spent fuel cladding.

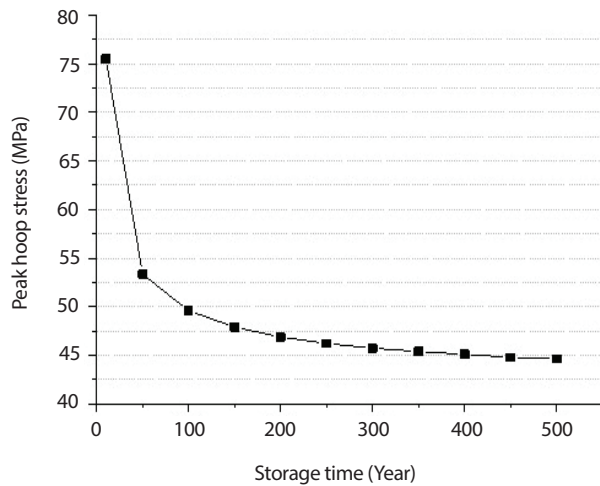


Fig. 5. Peak hoop stress of spent fuel cladding.

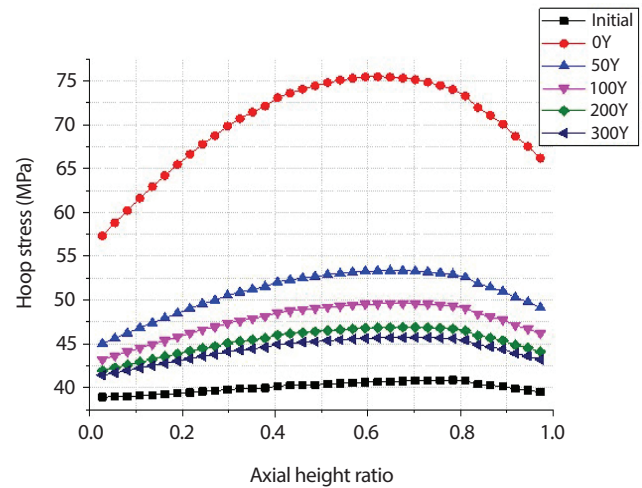


Fig. 6. Axial hoop stress distribution of spent fuel cladding.

equation was derived based on the result of the temperature distribution using the COBRA-SFS code, and added to the modified FRAPCON-4.0 code. As a result, the temperature of the spent fuel after the cooling time of 10 years in the storage pool was around 554 K. The temperature of the spent fuel showed a sharp change in the temperature from the beginning of the dry storage to 50 years, whereas rather gradual change was observed after 50 years. The temperature of the spent fuel after 300 years was observed to reach at nearly room temperature. Thus, this study focused on the results before 300 years. Fig. 3 shows the peak temperature of the spent fuel cladding with time, and Fig. 4 shows the axial temperature distribution of the spent fuel cladding with time.

3.2 Hoop stress prediction

The hoop stress distribution was predicted using the modified FRAPCON-4.0 code with Eq. (1) and the ideal gas equation. The hoop stress of the spent fuel after the cooling time of 10 years in the storage pool was around 75.5 MPa. The hoop stress of the spent fuel showed a sharp change in the hoop stress from the beginning of the dry storage to 50 years, whereas rather gradual change was observed after 50 years. The hoop stress of the spent fuel after 300 years appeared similar to the hoop stress distribution of the spent fuel discharged from the nuclear reactor. Fig. 5 shows the peak hoop stress of the spent fuel cladding with time, and Fig. 6 shows the axial hoop stress distribution of the spent fuel cladding with time. The results of the hoop stresses depended on the irradiation history and the cooling time.

3.3 Hydrogen concentration prediction

The hydrogen concentration distribution was predicted using the modified FRAPCON-4.0 code with Eq. (8) and numerical analysis. It showed that the axial hydrogen concentration increased as the burnup of the spent fuel increased. The peak concentration was approximately 600

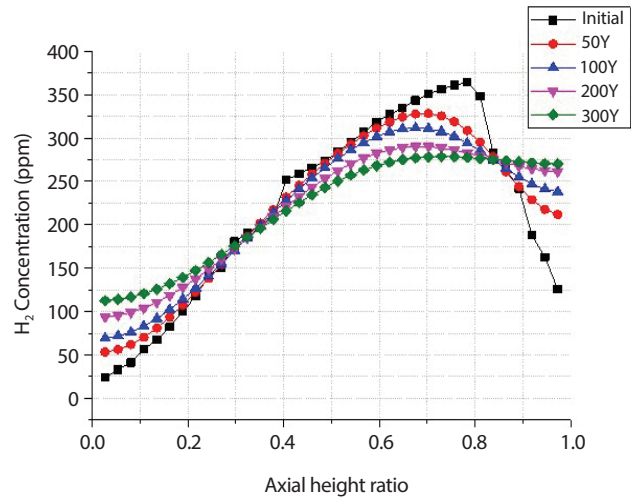


Fig. 7. Axial hydrogen concentration distribution through numerical analysis.

ppm, and the dissolution concentration was approximately 200 ppm. The hydrogen concentration in the fuel cladding was approximately 400 ppm. Hydrogen was transferred by concentration and temperature gradient over time, from an area with high concentration to an area with low concentration and from an area with high temperature to an area with low temperature. The initial temperature distribution and hydrogen concentration showed a parabolic line, where both temperature and hydrogen concentration above the axial center of the spent fuel were high but those at the both end of the spent fuel were low. As the dry storage period was extended, the hydrogen concentration above the axial center of the spent fuel decreased but that at the both end of the spent fuel increased. Fig. 7 presents the hydrogen concentration distribution on the dry storage condition through the numerical analysis.

4. Conclusions

This study predicted the axial cladding temperature, hoop stress, and hydrogen concentration of the spent fuel on a long-term dry storage condition, and developed a

program for the calculation processes. In addition, hydride effects of fuel cladding on a long-term dry storage condition were added to help users extend the range of application of the FRAPCON code.

The FRAPCON-4.0 code and the COBRA-SFS code were used to analyze the cladding temperature and hoop stress of the spent fuel. To predict accurately the axial and peak cladding temperature in the dry storage, the COBRA-SFS code was used to reflect the geometry of dry storage cask and heat transfer. The Westinghouse 15×15 fuel assembly and the TN-24P dry cask were applied. The cladding temperature was calculated using the COBRA-SFS code with the burnup information which was calculated using the FRAPCON-4.0 code. The FRAPCON-4.0 code was able to calculate the axial burnup distribution and the average burnup. The axial burnup distribution was the important factor to predict the axial cladding temperature because it depended on irradiation history. Spent fuel information such as rod internal pressure was used to calculate the hoop stress with ideal gas equation in dry storage.

The hydrogen concentration distribution of the spent fuel cladding depended on the axial power. The degree of hydrogen diffusion depended on the concentration and temperature gradient. So, the numerical analysis method was used to calculate the degree of hydrogen diffusion according to the concentration and temperature during the dry storage period.

Before 50 years of dry storage, the cladding temperature and hoop stress decreased rapidly. However, after 50 years, they decreased gradually and the cladding temperature was below 400 K. The spent fuel cladding during the dry storage period should be carefully managed because its temperature falls below the ductile-to-brittle transition temperature of the fuel cladding and it becomes vulnerable to impact. In addition, the hydrogen distribution showed that the concentration of hydrogen to the top and bottom parts of the spent fuel was higher as the storage period increased. The mechanical strength of the spent fuel cladding could be lower as the hydrogen concentration in the cladding

increased.

Initially, the United States planned to operate a dry storage facility for 20 years. But it was extended 40 years owing to the closure of final disposal project in the Yucca Mountain high-level radioactive waste repository. In addition, no specific decision was made on the future final disposal plan. Therefore, when either the final disposal of spent fuels after operation of the dry storage facility or the extension of the dry storage period is considered, the analysis of the characteristics behavior of spent fuels within the dry storage cask will provide the important information on the integrity of spent fuels.

Conflicts of interest

All contributing authors declare no conflicts of interest.

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