

Subsidy Project of Decommissioning and Contaminated Water Management

Upgrading of the Comprehensive Identification of Conditions inside Reactor

Accomplishment Report for FY2017

June 2018

International Research Institute for Nuclear Decommissioning (IRID) The Institute of Applied Energy (IAE)

All Rights Reserved. International Research Institute for Nuclear Decommissioning ©International Research Institute for Nuclear Decommissioning

Contents of the Report

- 1. Planning for the Project of Identifying of the Conditions inside the Reactors and the Project Structure
- 2. Project Achievements for FY2017
- 3. Research Details and Results Summary for FY2017



©International Research Institute for Nuclear Decommissioning

1-1. Project Needs

- For the decommissioning initiative for the Fukushima Daiichi Nuclear Power Station, it is crucial to estimate and understand the conditions of fuel debris and fission products (hereinafter FPs) inside the reactor pressure vessels (hereinafter RPVs). Direct observation inside the RPVs is still challenging due to the highly radioactive environment.
- A practical alternative measure is to comprehensively advance analysis and evaluation techniques to estimate the conditions inside the RPVs and the primary containment vessels (hereinafter PCVs) through accident progression analysis and evaluation of various measurement data and information, such as those collected onsite. This knowledge is expected to be used for the decommissioning process. To this end, initiatives for understanding the conditions inside the reactors have been pushed forward since FY2011, centering on improved severe accident analysis codes; the Modular Accident Analysis Program (MAAP) and the Severe Accident Analysis Code with Mechanistic (SAMPSON).
- Since FY2016, comprehensive activities have been carried out to advance identifying of the conditions inside the RPVs and PCVs. Not only the analytical results, using the accident progression analysis codes, but also plant parameters obtained at the time of the accident and other actual measurements will be used. Fact-based, on-site information collected through the internal PCV investigations, or dose investigations inside the R/Bs, in addition to the material science knowledge enhanced by recent studies will also be used.
- Amid such circumstances, the following technical development was conducted in FY2017.
 - In FY2016, the review was pushed forward with no improvement of the analysis codes based on the understanding of their capabilities and limits. In the end, it was necessary to upgrade the analysis codes to enhance the analysis results' reliability. Therefore, it was decided to make partial improvements to the analysis codes in FY2017.
 - Estimating the conditions inside the RPVs and PCVs by using the results obtained up to FY2016 were advanced. The study items selected for further research to reduce uncertainties of the estimated conditions inside the RPVs and PCVs were also analyzed and evaluated.



1-2. Application of Project Achievements and Project Target

- The aim of the project is to estimate the conditions inside the RPVs and PCVs through comprehensive analysis and evaluation of the study on issues to be solved. Specifically, major outputs of the project are shown in the below diagram; fuel debris and FP distributions that were estimated based on comprehensive analysis and evaluation. The diagrams were occasionally updated to reflect the latest results and provide them for other related research projects.
- In other words, the purpose of the project is to provide information that will be useful for fuel debris retrieval and decommissioning work.



1-3. Overall Plan: Implementation Schedule



IRIDAE

1-4. Project Overview and Structure for FY2017

O Project Overview

Identifying of the conditions inside the reactors has been sought by gathering knowledge from Japan and abroad and cooperating with overseas institutions. Accident progression analysis technology, analyzed actual data and on-site investigation results were used, as well as material science knowledge, etc. By using the highest level of technology available, the fuel debris and FP distributions inside the RPVs and PCVs as an input for the decommissioning task was estimated.

O Structure of the Project

This project is conducted under the cooperative framework between IRID and IAE, in which IRID manages and executes the overall project in cooperation with IAE. IRID conducts the project in cooperation with the IRID member organizations, which are JAEA (CLADS), Toshiba Energy Systems & Solutions, and Hitachi-GE Nuclear Energy. In addition, the project was promoted by closely sharing information with relevant organizations, such as NDF and TEPCO, and thereby obtaining cooperation from them.



Core team: An organization consisting of project participating organizations. Responsible for project policymaking and estimating conditions inside the RPVs and PCVs.



1-5. Implementation Items and Schedule for FY2017

6

	First half of FY2017			S	Second half of FY2017		
(1) Comprehensively analyze and evaluate the conditions inside the reactors							
(1) Comprehensively analyze and evaluate based on the Units data and other R&D project results		Present study ite	ms for further	research by using da	atabase functions		
[1]-1 Extract issues for further research to reduce uncertainties of estimated conditions inside the reactors	Extract issues for furt	ther research by rev	iewing the FY2	2016 results, measure	ement data, and on-site inspecti	on results	
	Und	lerstand the condition	ons inside the	RPVs and PCVs and	estimate the fuel debris and FP	distributions	
[1]-2 Comprehensively analyze and evaluate based on the Units data and other R&D project results		Review	1	Summarize	Review	Sumr	
R&D project results	Other project results, test data, etc.						
	c.	Collect an	d organize informatio	on			
[2] Establish a database for comprehensive analysis and evaluation							
			Enhance an	d improve the DB			
(2) Estimate and evaluate fuel debris behavior and fission product behavior and their characteristics for comprehensive analysis and evaluation							
[1] Reduce uncertainties by using analytical techniques, etc.	Preparation			Analyze the accider	nt scenario		
[1] Reduce differrainties by using analytical techniques, etc.	Evaluate the analysis codes						
	Analyze based on the plant data at the time of the accident Enhance necessary tests and data, and verify the analysis model						
	Preparation						
[2] Evaluate FPs' chemical characteristics				Analyze the acciden	t scenario		
	Evaluate the analysis codes						
		Enl	hance necessa	ary tests and data, an	d verify the analysis model		
			В	SAF Phase-2			
[3] International joint research to use knowledge inside and outside Japan							

2. Project Achievements for FY2017



2-1. Overall Structure of the Project for Identifying the Conditions inside the Reactors

- (1) Comprehensively analyze and evaluate the conditions inside the reactors
- [1] Comprehensively analyze and evaluate based on the Units data and other R&D project results
 - 1 Extraction of issues for further research to reduce uncertainties about the estimated conditions inside the reactors
 - 2 Comprehensive analysis and evaluation based on the Unit data and other research development results
- [2] Establishment of a database for comprehensive analysis and evaluation (IAE)
- (2) Estimation and evaluation of fuel debris behavior, and fission product behavior/characteristics toward comprehensive analysis and evaluation
 - [1] Reduce uncertainties by using analytical techniques, etc.
 - 1 Evaluation of the concrete erosion at the pedestal using sensitivity analysis (Hitachi GE)
 2 Inverse problem evaluation using virtual reactors and a compiled database (JAEA)
 3-(a)-1 Estimation of the amount of fuel debris that remains in the pedestal space obstacles (IAE)
 3-(a)-2 Detailed evaluation of debris behavior inside the post-damage lower head (IAE)
 3-(a)-3 Evaluation of the condensation behavior of water vapor containing hydrogen in the S/P (IAE)
 3-(a)-4 Event sequence analysis at the time of core material slumping (JAEA)
 3-(a)-5 Three-dimensional evaluation of MCCI behavior (IAE)
 3-(a)-6 Determination of factors influencing debris distribution (IAE)
 3-(a)-7 Analysis of each unit over three weeks after the accident (IAE)
 3-(a)-8 Detailed accident progression analysis using accident progression analysis codes (Toshiba Energy Systems & Solutions)
 - 6 Simulated fuel assembly degradation test (JAEA)

IRID

8

2-1. Overall Structure of the Project for Identifying the Conditions inside the Reactors

- (2) Estimation and evaluation of fuel debris behavior, and fission product behavior/characteristics toward comprehensive analysis and evaluation
 - [2] Evaluation of FPs' chemical characteristics (JAEA, IAE)
 - 1 Reaction between cesium and steel materials, and re-evaporation
 - 2 Evaluation of particulate cesium compounds
 - 3 Optimization of the cesium compounds evaluation model
 - 4 Analysis and evaluation of the units using the upgraded model
 - 5 Analysis of samples collected at Fukushima Daiichi Nuclear Power Station (NPS)
 - [3] International joint research to make use of knowledge inside and outside Japan (IAE)



9





IRIDIAE

2-4. Results of Comprehensive Analysis and Evaluation





IRIDIAE



IRIDAE



IRID



IRID



IRIDAE



The dash-dotted line shows the inside of the containment is asymmetrical with the pedestal.

IRIDIAE









IRIDA

Estimate Diagram of Debris Distribution and the RPV and Legends PCV Conditions in Unit 3 • From an estimate energy amount based on the PCV pressure increase caused by the generated hydrogen, it is presumed that • At present, both possibilities exist for the shroud: it could be sound or damaged. • Oxide debri

- most of the fuel melted. (actual measurement and analysis) There was no temperature increase in each section of the RPV when the CS system was stopped from December 9 through 24, 2013 (the flow rate from the FDW was increased, and the total volume of water injection had remained stable). Based on this finding, there should be little fuel debris in the reactor core (less than that in Unit 2). (actual measurement)
- In addition to the above finding, the temperature at the lower part of the RPV decreased when the total water injection volume increased as water injection from the CS system began (on Sep 1, 2011); hence, fuel debris is probably in the lower plenum. (actual measurement)
- The muon measurement results indicate there may be no large mass of fuel debris in the original reactor core area. (actual measurement)
- Because a structure, seemingly CRGT, fell out of the pressure vessel, it is presumed there is a rupture large enough for the CRGT to fall through. (actual measurement)
- Inside the pedestal, there were water surface fluctuations in the center and periphery of the RPV; therefore, ruptures may exist in the center and periphery of the RPV. (actual measurement)
- Since there is a gap between the flange faces at the lower part of the CRD housing, part of the welds at the CRD housing and the pressure vessel bottom may not be firmly fixed. (presumption based on the actual measurement)
- The internal PCV investigation revealed that the damage inside the pedestal had progressed further than in Unit 2, suggesting that the unit's PCV has more dropped fuel debris than Unit 2's. (actual measurement)
- Damage to the platform is presumably due to the drop of high-temperature debris. (actual measurement)
- Damage and depositss (the latter seemingly a result of solidified molten material solidification) were observed on the metal fittings to support the CRD housing; fuel debris may exist above and below it and also in its surroundings. (actual measurement)
- The lower part of the pedestal has some substances, seemingly a result of molten material solidification, and other dropped objects such as grating and depositss. (actual measurement)
- A pool of water on the PCV floor, if any, may have formed particulate debris. (general assumption)
- Particulate debris, if any, is likely to be found in a stagnant spot. (general assumption)

IRIDIAE



- Legends Remaining fuel rod and its debris Oxide debris (porous) (general assumption and analysis) Particulate debris Since the increase in fuel temperature might not be so great in the peripheral area, fuel rod debris and Fuel debris (rich in metals)* pellets may still exist there. (general assumption, testing, and analysis) Even if some fuel rods are present, they should be Concrete mixed debris just a small portion of the peripheral area. (general assumption) It is estimated to be general oxide debris from molten CRGT fuel solidification, (general assumption) Damaged CRGT Part of the CRGT remains un-melted when there is a little heat transfer from high-temperature molten CRD debris. (general assumption) If particulate debris or pellets are present, they may CRD (with debris inside) accumulate in a stagnant spot. (general assumption) Shroud The muon measurement results suggested that part of the fuel debris still may exist at the bottom of the Damaged shroud* RPV; however, the possibility is uncertain. (actual measurement) 23 Pellet As a result of damage to the CRGT and CRD housing, a small amount of fuel debris or molten metal may have entered the CRD housing. (general assumption and Ruptured hole of the RPV testing) Upper tie plate* Part of fuel debris may have solidified without MCCI. (general assumption) deposits (material unknown)* Considering that D/W was sprayed for more than an hour, from 7:39 on March 13 as part of the accident response, water may have accumulated on the D/W Ballooning fuel* floor when the pressure vessel suffered damage, and it may have prevented fuel debris from spreading Oxide debris* further. (actual measurement and general assumption) Fuel debris spread out of the pedestal through the pedestal opening, but presumably no shell attack leavy metal debris* occurred. (actual measurement and analysis) Particulate pellet* Cladding residue* An explosion occurred in Unit 4
 - and then in Unit 3, possibly due to hydrogen generated by MCCI. (actual measurement)

The dash-dotted line shows the inside of the containment is asymmetrical with the pedestal. * Not used in the estimate diagram of Unit 3



Molten reactor internals*

Control rod mixed melt*

Solidified B4C*

Unit 3: Estimate Diagram of *The estimate was made focusing on cesium, a major source nuclide **FP** Distribution <Chemical forms and characteristics of major types of cesium estimated> The gamma-ray spectra measurement on the operation floor in Unit 3 Cesium iodide, Cesium hydroxide, Cesium chloride •Easily evaporate and tend to escape from the pressure vessel as vapor due to a indicated high dose levels in the shield plugs' gaps and joints. The difference in pressure or concentration. pictures at the time of the accident also show a large amount of vapor •Have a hydration property and tend to move with moisture by, for example, water was discharged from the damaged reactor building. vapor condensation or dew condensation (on the wall surface). Therefore, it is presumed that FPs were discharged through the Cesium molybdate, Boric acid cesium following route: pressure vessel -> containment -> containment top Have a lower vapor pressure compared to the afore-mentioned substances and, head flange -> reactor well -> shield plug -> operation floor. It was also hence, tend to stay inside a pressure vessel. figured that FPs exist unevenly along the FP migration route. Cesium molybdate may form because of a reaction between cesium and steel material containing molybdenum. Compound of silicon and cesium Hydrated cesium compounds may have penetrated - Insoluble cesium particles (amorphous particles including cesium with its through the concrete surface in the form of cesium main component of silicon oxide) hydroxide. Believed to be difficult to hydrate, and particles the size of a few microns tend to soar up. - Products of reaction with steel material (crystalline substances) Although the fuel melt caused a high temperature Believed to be difficult to hydrate, and tend to stay in the oxide layer on the steel inside the pressure vessel, the separator and dryer are surface when in the 800-1,000° C temperature range. highly likely to have retained their shapes, according to the muon measurement results of Unit 3. Depending on the conditions of the gas phase, such as the water vapor/hydrogen ratio, cesium molybdate or boric acid cesium may have The level of activity at 0.7 m below the surface of the served as major chemical species, suppressing their discharge to the stagnant water in the PCV was lower than that near the outside of the pressure vessel. water surface. The specimens collected at the two locations near the water surface of the stagnant water in the PCV, and about 0.7 m below the water surface, contained Cs134, In the early stage of the core heating, insoluble cesium particles Cs137, tritium, and Sr90.(Oct 30, 2015) were generated by coagulation reaction between silicon oxide and Cs134 concentration [Bq/cm³]: 4.0E + 2 (near the water cesium hydroxide in the gas phase; however, it was estimated that surface), 2.3E + 2 [7] (approx. 0.7 m below the water surface) the amount generated is smaller than that in Unit 2. Cs137 concentration [Bg/cm³]: 1.6E + 3 (near the water surface), 9.4E + 2 [7] (approx. 0.7 m below the water surface) Depending on the temperature/atmosphere history, some cesium in the vapor phase inside the pressure vessel is thought to be taken into the oxide layer of the steel material due to the reaction with silicon oxide on the steam-oxidized steel surface. Considering this factor, the high radiation may have occurred at the separator and dryer, which have large surface areas. The FPs that migrated to the S/C and got stuck in Cesium possibly remains inside the the pooled water may still exist in the stagnant · FPs may unevenly exist in fuel debris. fuel debris although its amount is water. expected to be small. • A huge amount of FPs may still exist, given the large volume of stagnant water in the S/C.

IRID



IRID

©International Research Institute for Nuclear Decommissioning

(1)-[2] Establish a Database Necessary for Comprehensive Analysis and Evaluation

[Overview and Objectives]

•Promote the enrichment of the database by maintaining and managing it and updating its contents in a continuous and timely manner so that the latest measurement data and research results can be viewed, analyzed, and downloaded from the database. Also, upgrade the display functions to present the analysis results in a more user-friendly manner.

•The website for the OECD/NEA BSAF project (English site) is run separately; however, since the Japanese and English sites have a lot of information in common, the two sites will be managed in an integrated manner. By doing so, information sharing in English will be enhanced.

[FY2017 Research Details]

- Improve the operability of the graph display functions (including the zoom function)
- Integrated management of the English and Japanese databases
- · Timely update and management of the contents

[Final Report]

• Detailed display range settings for the graph display function were introduced. A display selection function to provide an easy-to-view display of the measurement data collected within the first three weeks after the accident was also introduced.

•The English version of the document search function to enable the search of English documents on the internal PCV investigations, etc., was developed.

•The timely update and management of the database contents was introduced.

•By implementing the afore-mentioned measures, making the comprehensive analysis and evaluation mentioned in (1)-[1] more efficient was contributed to.

[Database URL] https://fdada.info/

IRIDIAE

Unit	01 02	2 03 04	4 ⊞5	6			
Data type	Photo Accumulated water Water level Gamma camera TMI-2 Inside PCV Torus room Tm			Dose rate Video FP concentration Muon measurement Robot investigation Core sampling			
Location				Inside RPV TIP room Exhaust stack			
Release year	© 2011 ≷ 2017	© 2012 ⊮ 2018	2013	8 🗏 2014	© 2015		
Free word	Enter syar	ch string			Search Clea		
Title			ЯĽ	Website	Date		
♥ Contains_		_	Vic	\ ♥ Contain	Contains.		
Videos to show 5 Primary Cont (Reference vide investigation)	ainment Ve	sel (PCV)		TEPCO	2017/02/27		

Figure: Developing the English version of Document Search

(2)-[1]-1 Sensitivity Analysis of the Concrete Erosion at the Pedestal

[Overview and Objectives]

Based on the results of the investigation inside Unit 1, a sensitivity analysis of MCCI was conducted to help estimate the fuel debris conditions in the pedestal.

[FY2017 Research Details]

- Since the internal PCV investigations indicate that the pedestal is standing on its own, the key parameters that affect concrete erosion using the MAAP codes were studied.
- The significant effect of a concrete melting when no water was injected from a fire engine was verified and reflected the finding in the analysis conditions for long-term erosion.
- By using three-dimensional codes, long-term erosion analysis was performed to consider the differences in the falls of fuel debris and their resultant deposits formulation.

[Final Results]

RIDIAE

- The pedestal erosion tendency for each fuel debris fall location and deposits state was grasped. The tendencies indicated the possibility that, even in a situation where no water is injected from a fire engine, no major pedestal erosion occurs, although it may depend on the concrete's properties.
- The sensitivity analysis results were reviewed, including those obtained up until the last fiscal year, and reflected the pedestal erosion tendencies in the fuel debris distribution diagram.







Figure 2. Temporal change in fuel debris temperature and structure material temperature (Case 2)

(2)-[1]-1 Sensitivity Analysis of the Concrete Erosion at the Pedestal

[Application to the estimate diagram of fuel debris distribution] Fuel debris shape and concrete erosion behavior





- Even if fuel debris depositss solidify, they may remelt because of decaying heat and flatten.
- It is possible that no major pedestal erosion occurs, even without water injection from a fire engine, although it depends on the concrete's melting point. (Case 1 and 3 showed the same tendency.)

 Molten debris
 Concrete floor

 500
 1,000
 1,500
 2,000
 2,500

 Crust
 Concrete outside the shell
 Temperature (K)





Eroded area (concrete -> fuel debris) Pedestal wall/base and PCV shell

Figure 3. Case of eccentric fall and depositsion (Case 2)

Figure 4. Review on the damage condition of the pedestal base and wall (Case 2)



28 (2)-[1]-2 Inverse Problem Evaluation Using Virtual Reactors and Compilation of a Database

[Overview and Objectives]

From right after the accident until today, huge and various kinds of data were released to the public, including the temperature and pressure data inside the PCV. By solving inverse problems from those data, fuel debris locations are estimated. [FY2017 Research Details]

Virtual reactors were created by making use of three-dimensional CFD codes (StarCCM+). In addition to the creating more RPV models, the temperature distribution from one month to six months after the accident was evaluated, with the forward problem analysis approach, and the fuel debris locations inside the plant was estimated.

[Final Results]

[Unit 1]

temperature

distribution

Almost no heat source inside the RPV is expected. Meanwhile, given the leakage of superheating vapor from the RPV near the safety valve, the presence of a heat source in the RPV needs to be assumed.

[Unit 2]

For an assumed situation where most of the heat source exists in the RPV and some 0.8MW (about 25% of the estimated total heat generated) is present in the pedestal, the temperature distributions in the steady and transient states were successfully simulated.

[Unit 3]

The temperature distribution suggested the possibility that the heat source of about 0.25-0.3 MW (about 25–30% of the estimated total heat generated) is present in the lower part of the RPV. According to a calculation, a significant amount of heat, which is estimated to be 0.8MW (81% of the estimated total heat generated) or greater, was removed by feedwater.



Estimated heat source distribution Apr 6, 2011 Estimated





(2)-[1]-3-(a)-1 Estimate of the Amount of Fuel Debris Remaining in the Pedestal Space Obstacles

[Overview and Objectives]

Fuel debris may have attached to structures, such as the grating and the penetration pipe, in the pedestal space. From the Unit 2 internal PCV investigation results, thermal balance data, etc. the amount of fuel debris stuck in those obstacles are estimated.

[FY2017 Research Details]

Through comprehensive analysis of temperature distribution and temperature behavior upon changes in the water injection operation, the presence or absence of fuel debris in the pedestal space was evaluated. A heat transfer analysis was also conducted by referring to the results of the internal PCV investigation held in January and February 2017 and to the accident scenario, and how much fuel debris may have got stuck in the structures near the CRD housing was evaluated. [Final Report]

Since the temperature in the lower part of the CRD housing is almost the same as that of the D/W (Figure 1), it is estimated that there are no large amount of fuel debris near the CRD housing. Possible locations on the structures where fuel debris may have got stuck (Figure 2) were extracted and a heat transfer analysis (Figure 3) was performed. The results indicate the possibility that the structures did not melt and that a small amount of fuel debris still exists.





(2)-[1]-3-(a)-2 Detailed Evaluation of Debris Behavior inside the Lower Head after Damage of the Lower Head

[Overview and Objectives]

It is important to find out whether fuel debris still exists inside the CRD housing in the lower part of the RPV for debris removal purposes.

[FY2017 Research Details]

Based on the specimen cutting inspection in the CRD housing melting test, the model parameter was optimized and the fuel debris behavior in the lower head (Figure 1) was analyzed. To evaluate the presence of fuel debris near the CRD and based on the KAERI test results, a sensitivity analysis was performed with the detailed meshes showing fuel debris temperatures, the presence/absence of water in the lower head, decay heat, etc. (Figure 2 and 3).

[Final Report]

The amount of fuel debris penetrating the CRD housing largely depends on the fuel debris temperature when melting occurred in the upper part of the CRD housing and, hence, involves significant uncertainties. The detailed analysis results showed that a high temperature persisted near the weld, even when water was present in the lower head, indicating that fuel debris may have flowed out through the gap under the weld of the RPV and CRD housing.



IRIDIAE

30

(2)-[1]-3-(a)-3 Evaluation of Condensation Behavior of Water Vapor Containing Hydrogen in the S/P

[Overview and Objectives]

Vapor condensation behavior (a phenomenon of incomplete condensation) in the S/C is important in identifying the accident progression. Especially, the behavior of mixed gas of hydrogen and vapor blowing into the S/C is important for evaluating the PCV/RPV pressures. Through evaluating this behavior, and projecting the amount of generated hydrogen, etc., a reduction in the the uncertainties of fuel debris migration behavior is sought.

[FY2017 Research Details]

By combining SAMPSON and the CFD tool POOL-3D with the temperature distribution and the condensation experiment on steam containing non-condensable gas (LINX), the condensation behavior of water vapor containing hydrogen in the S/C is studied. Based on the study results, the distribution of S/C water temperatures are provided, a key parameter regarding condensation behavior, in the three-week analysis "(2)-[1]-3-(a)-7."

[Final Report]

The three-dimensional temperature distribution inside the S/C during the period in which vapor and hydrogen were discharged from the SRV, was evaluated. It was found that, in Unit 2, there was a temperature difference of approx. 30° C up until the RCIC stopped (70 hours after the scram). After forced depressurization (75 hours after the scram), the temperatures inside the S/C equalized; however, it was found that temperature stratification occurred again due to vapor and hydrogen erupting from the SRV. It was found that, in Unit 3, temperature stratification occurred while the RCIC was in operation and it persisted while the HPCI was working. It as also found that, after the HPCI stopped, temperature stratification was resolved by a huge amount of hydrogen generated during the progression of core melt.



Figure 1. Unit 2 temperature stratification at indicative event



Figure 2. Unit 3 temperature stratification at indicative event



31

2)-[1]-3-(a)-4 Event Sequence Analysis at the Time of Core Material Slumping

[Overview and Objectives]

In all units of Fukushima Daiichi NPS, the core materials are believed to have relocated to the lower plenum (slumping) and to the pedestal. The core state during the slumping affects the RPV failure mode and has a significant impact on the eventual material distribution and their properties. The corresponding accident progression is therefore determined by conducting the SA code analysis and using the plant data (such as pressure) to improve the accuracy of the evaluation of the final materials distribution and their properties.

[FY2017 Research Details]

A detailed MELCOR code analysis (Waseda Univ.) of the accident progression in Units 2 and 3 was performed, with a focus on the consistency with the plant data. The most probable transitions and uncertainties was also evaluated by comparing the results with the MAAP (EPRI), SAMPSON (IAE) and SCDAPSIM (JAEA) results all the while shedding light on core energy.

[Final Results]

The following knowledge was obtained and provided in the estimate diagram of fuel debris distribution.

- Based on the evaluation of core energy, it was concluded that most of core fuel did not melt when slumping occurred in Unit 2 (the best estimate). It was concluded, meanwhile, that in Unit 3, where it took a long time from the initiation of core damage to slumping, a considerable amount of fuel melted due to accumulated energy (the best estimate) (Figure 1).
- Based on the energy state described above, the effect on the heat transfer behavior of fuel debris and structural materials in the lower plenum (Figure 2) was assessed and a result matching the internal investigation's results was obtained.

Reflection in the estimate diagram of fuel debris distribution

The combined knowledge with the results of the "simulated fuel assembly degradation test" indicates a considerable amount of un-melted fuel pellets may still exist in the reactor core or at the lower plenum area in Unit 2. The knowledge indicated a relatively large mass of solidified fuel may still exist in the pedestal or CRD area in Unit 3. The Unit 3 data analysis results also indicate the possibility that MCCI was mitigated by the water in the PCV.



Figure 1. Comparison of the accumulated core energy (the best estimate) and timing of core material migration to the lower plenum and pedestal in Units 2 and 3



©International Research Institute for Nuclear Decommissioning Figure 2. Effect on the heat transfer behavior of fuel debris and structure material estimated according to the energy during slumping

(2)-[1]-3-(a)-5 Three-Dimensional Evaluation of MCCI Behavior

[Overview and Objectives]

A quantitative evaluation of the state of concrete erosion caused by MCCI that is assumed to have occurred in the pedestal was performed.

[FY2017 Research Details]

In the last fiscal year, a sensitivity analysis on Unit 1 was conducted, finding that the volume and temperature of dropped fuel debris are the key parameters of fuel debris diffusion. In this fiscal year, with regard to Units 2 and 3, a three-dimensional evaluation on the MCCI behavior under a condition where water remains on the pedestal floor was performed. A sensitivity analysis using parameters such as the remaining water volume was also conducted.

[Final Report]

- It was verified that, in Unit 2, the whole fuel debris would remain in the pedestal if the dropped debris was 20 tons (12.8 tons of fuel debris) or 40 tons (25.6 tons of fuel debris) and concrete erosion stopped within 10 hours.
- With regard to Unit 3, an analysis for a condition where the dropped debris was 140 tons (89.4 tons of fuel debris) was performed, by setting the sensitivity parameters at the initial water level and the drop velocity of fuel debris. The results of the analysis with SAMPSON/DSA showed the initial water level had a limited impact on the diffusion of fuel debris, while the drop velocity of debris had a large impact on fuel debris diffusion.





(2)-[1]-3-(a)-6 Confrimation of Factors Influencing Debris Distribution

[Overview and Objectives]

A quantitative evaluation of the effect on fuel debris distribution is performed, assuming the fuel rods collapsed before melting, to ensure reliability of the fuel debris distribution information that could be obtained through the accident progression analysis, thus reducing uncertainties.

[FY2017 Research Details]

A model representing the pre-melt collapse of fuel rods was introduced and a quantitative evaluation on the effect of this phenomenon on the fuel debris distribution was conducted. The fuel debris distribution was comprehensively evaluated while taking into account the results of the two-dimensional temperature distribution evaluation that studied the output per fuel assembly.

[Final Report]

- In the VERCORS test, a fuel rod collapsed at approx. 2,600 K, meaning that it would take about 100 hours for a Larson-Miller parameter model to collapse when the fuel rod temperature is around 2,300 K. Therefore such factors as the loss of metal cladding, due to melting and oxidation, and the vibration of fuel rods, due to the sudden generation of vapor, were taken into account and a model using the fuel rod temperature and time history for the SAMPSON code was introduced to simulate a fuel rod that collapses within a few hours even at 2,300 K.
- To evaluate the possibility of localized collapse of a fuel rod that cannot be evaluated through the SAMPSON analysis, the temperature distribution at the horizontal section of Unit 2's core was evaluated. For the latter evaluation, factors such as radiation and the Zr-water vapor reaction were taken into account, getting the following results.

Evaluation of the two-dimensional temperature distribution based on the SAMPSON three-week analysis

- 1. The fuel rod temperature increased as shown in the figure; however, it is more likely that the supporting strength in the lower part of the fuel rods was lost before the fuel failure occurred.
- 2. When the emissivity and the amount of hydrogen generation to an extent consistent with the knowledge and actual measurements in the past was changed, a result in which part of the fuel at the outermost ring collapsed was gotten.

Presumably the support structure at the lower part of a fuel rod starts losing its strength before the fuel rod starts collapsing.

Also presumably, a fuel rod collapses downwards when the fuel falls toward the center; or collapses onto the shroud when the fuel falls toward the shroud.



Evaluation results of two-dimensional temperature distribution (at 82 hrs)



(2)-[1]-3-(a)-7 Analysis of Each Unit over Three Weeks after the Accident

[Overview and Objectives]

Analysis was performed for up to three weeks, by which time the MCCI was expected to have reached a completely steady state, in order to evaluate the final fuel debris distribution all the while taking uncertainties into account.

[FY2017 Research Details]

Sensitivity parameters affecting the distribution, composition, and properties of fuel debris were extracted and a sensitivity analysis was performed. A three-week analysis was performed with each sensitivity parameter group that matches the actual measurements obtained from the units and the results of the on-site inspections including the internal PCV investigations. [Final Report]

- To reduce uncertainties in the accident progression scenario, which is the prerequisite for evaluating fuel debris migration behavior and the final fuel debris distribution, accident progression scenarios to the most likely ones were narrowed down. This process was completed by comprehensively evaluating results of studies conducted inside and outside Japan, such as internal investigations using muon and robots, measurements of pressure and water levels in the reactors, the knowledge and scientific considerations gained or made until today, and BSAF.
- This initiative was conducted by IAE, the implementation organization for this particular study item, and in cooperation with the parties involved in the Project (JAEA, Hitachi GE, Toshiba, and TEPCO) through discussions at study meetings (which were held six times).
- The SAMPSON analysis results, which were obtained based on this accident progression scenario, matched well with the actual measurement results, significantly contributing to reducing the uncertainties for the final evaluation of the fuel debris distribution.



RPV pressure behavior (Unit 2)



RPV pressure behavior (Unit 3)

35





©International Research Institute for Nuclear Decommissioning
(2)-[1]-3-(a)-7 Analysis of Each Unit over Three Weeks after the Accident

[Final report (evaluation results of Unit 1)]

- From the discussions made at the accident-scenario study meetings and the SAMPSON analysis results, it was presumed that the fuel rods started fracturing about 4 hours after the scram and started melting in about 5 hours. Based on these presumptions, fuel debris fell on the lower plenum in about 11 hours and further down to the pedestal in about 15 hours. It was concluded, therefore, that the fuel rods and fuel debris (containing fuel and structure materials) stayed in the reactor core area for about 5 hours in the high-temperature state due to decay heat and oxidation reaction.
- The VERCORS and CMMR tests indicate the possibility that the fuel rods maintain their column shape up to around 2,600 K. Considering the melting point of the channel box covering the fuel rods is 2,100 K (or 1,500K or even lower due to eutectic formation), the shroud would be directly exposed to radiation from fuel rods when their temperature far exceeds the melting point of 1,700 K. For Unit 1, no water injection was conducted while core damage progressed; therefore, it was concluded that the water in the down-comer may have fallen to the BAF or lower in 5 to 7 hours after the scram.
- The SAMPSON analysis reached the same conclusion, indicating that the shroud would fracture significantly and melt. Considering the phenomenon in which a shroud loses its structural strength at around 1,000 K before it melts, it was concluded that the shroud is deformed, damaged or buckled by the weight of the structures at the upper portion (such as the separator).
- It was also concluded that the structures in the lower plenum (such as the CRGT) are damaged or melted further than their counterparts in Units 2 and 3. It was also presumed that the concrete erosion by MCCI, even the largest one, will stop near the PCV shell.



IRID

©International Research Institute for Nuclear Decommissioning

(2)-[1]-3-(a)-7 Analysis of Each Unit over Three Weeks after the Accident

[Final report (evaluation results of Unit 2)]

- From the discussions made at the accident-scenario study meetings and the SAMPSON analysis results, it was concluded that the fuel rods started fracturing about 77 hours after the scram and started melting in about 78 hours. Then, it was presumed that about 100 tons of fuel debris (containing fuel and structure material) fell on the lower plenum in about 80 hours and some part of fuel debris fell further down to the pedestal in about 92 hours.
- The PCV pressure presumably rose between Hour 80 and 81, generating hydrogen and accelerating damage to, and melting of, the fuel rods. The LINX test results indicate the possibility that non-condensable gases, such as hydrogen, inhibit the condensation of water vapor. As is shown by actual values, the S/C water temperature rose to a high level during the hour, indicating our need to take incomplete condensation into account when evaluating the amount of hydrogen generated.
- A SAMPSON analysis was performed that took account of the actual measurement data and the phenomenon, getting a result showing that at least nearly 300 kg of hydrogen was generated. The analysis results showed that peripheral fuel rods may not melt completely under some conditions.
- A two-dimensional analysis ((2)-[1]-3-(a)-6) of the in-core temperature distribution was conducted, taking into account such factors as the oxidation reaction heat resulting from hydrogen generation, water injection and vapor temperature that was obtained from the SAMPSON analysis, to closely evaluate the fuel rod temperatures. A presumption was reached that the support structure of the peripheral fuel rods nearly rose to the melting point and that almost all of them eventually collapsed. However, some fuel rods may still exist, leaning on the shroud side.





©International Research Institute for Nuclear Decommissioning

(2)-[1]-3-(a)-7 Analysis of Each Unit over Three Weeks after the Accident

38

[Final report (evaluation results of Unit 3)]

- From the discussions made at the accident-scenario study meetings and the SAMPSON analysis results, it was concluded that fuel debris (fuel and structure material) slumping occurred at 43 hours and 45 hours after the scram. It was concluded that the PCV pressure continued to rise after the PCV vent closed, but that increase stopped as a result of balancing with the leakage from the D/W. Given the fact that the water injection from the fire engine was limited, it was concluded that the PCV pressure finally started decreasing when the inside of the RPV dried out. It was concluded that, during that period, water started migrating from the S/C to the D/W, creating, along with the water due to the D/W spray, some pools of water on the D/W floor.
- It was concluded that the lower head damaged 50 hours after the scram and that fuel debris dropped to the pedestal. From the
 analysis results showing almost zero residue of fuel debris in the reactor core, it was concluded there are about 30 tons of
 crusted fuel debris in the lower plenum and about 120 tons of fuel debris in the pedestal.
- A relatively large amount of water was present when the fuel debris migrated to the pedestal. Considering the possibility that water seeped in the cracks of the crust on the surface, it was concluded that the fuel debris cooled down quickly, resulting in almost zero concrete erosion.



(2)-[1]-3-(a)-8 Detailed Accident Progression Analysis Using the Accident Progression Analysis Codes

[Overview and Objectives]

- In the detailed accident progression analysis conducted in FY2016 by using the MAAP code, the accident progression scenario and the fuel debris distribution was studied while taking into account the code's limit and for a physical phenomenon model. The issues of reduction and quantification of the fuel debris distribution were also extracted.
- In FY2017, some of the physical phenomena models of the code were changed. The renovated models in the accident
 progression analysis were used to reduce the uncertainties involved in estimating the accident progression scenario and the
 fuel debris distribution.

[FY2017 Research Details]

- Improvement of the MAAP physical phenomena models
 - Model of fuel debris outflow when the RPV was damaged
 - Model of B₄C oxidation of the residual heat control rods
 - > Model of core blockage and corium migration
 - Model of fuel debris flowing into the CRD housing
- Detailed evaluation of fuel debris dispersion behavior for the study on MAAP boundary conditions
- Estimation of the accident scenario and quantification of the fuel debris distribution based on the accident progression analysis on Units 2 and 3 using the improved code

[Final Results]

- Improved the MAAP physical phenomena models and testing its operation and validity
- Performed the detailed accident progression analysis in Units 2 and 3 using the improved code
- Updated the accident progression scenario and the estimated fuel debris distribution



Example of model improvement (core blockage and corium migration model)

The model initially did not return to Type 2 or 3 once it reached the blockage node (Type 4); a core damage progression model was made allowing such return in order to simulate the accident that presumably occurred at Fukushima Daiichi. The impact on the amount of hydrogen generated in the core damage process was particularly significant, and the reproducibility of the PCV pressure behavior was improved.



(2)-[1]-3-(a)-8 Detailed Accident Progression Analysis Using the Accident Progression Analysis Codes

Unit 2

Area	Results of improved MAAP analysis	Estimated fuel debris distribution based on the analysis results
Reactor core area	In the reactor core, 14.5% of the initial UO_2 mass still exists.	Most parts of the reactor core fell to the lower plenum. The outermost peripheral area of the reactor core with low output may still exist on the core support plate because of the long-term water injection from the fire engine.
Lower plenum	In the lower plenum, 65% of the initial UO_2 mass still exists as particulate fuel debris.	Due to the localized damage to the weld of the instrumentation guide tube, and solidification on the RPV sub-structure surfaces, the molten core may have gradually fallen to the lower part of the PCV. A rupture may exist at a location closer to the side of the RPV boom than to the center, possibly limiting the fall of the molten core only from a position at or higher than the height of the rupture.
Lower part of the PCV	Of the initial mass of the reactor core, 20% falls to the pedestal.	At around 14:00 on March 15, the molten core, presumably part of the molten core in the lower plenum, possibly fell to the lower portion of the PCV. Concrete erosion may have occurred to a very limited extent.

Unit 3

Area	Results of improved MAAP analysis	Estimated fuel debris distribution based on the analysis results
Reactor core area	Fuel no longer exists in the reactor core.	Most parts of the molten core may have fallen, in a large scale, from the reactor core area to the lower plenum; as a result, the amount of the fuel that still exists in the reactor core area may be limited.
Lower plenum	Of the initial mass of the reactor core, 20% still exists in the lower plenum.	Due to the major damage at the RPV bottom, the amount of the molten core that still exists in the lower plenum may be small.
Lower part of the PCV	Of the initial rector core mass, 80% fell to the pedestal and then part of it flowed into the drywell.	Due to the massive molten core falling to the lower part of the PCV, the molten core may have flowed from the pedestal to the drywell, but in a limited manner. There may be more concrete erosion toward the floor.



(2)-[1]-6 Simulated Fuel Assembly Degradation Test

[Overview and Objectives]

Regarding the core melting and migration behavior at the time of the accident on the 1st floor, it might have had a residual, heat-type progression, different from the TMI-type. Factors such as (1) gas permeability in the high-temperature core, and (2) entry of decayed fuel into the lower support structure area substantially affect the presence or absence of molten fuel in the core. These have greatly contributed to the uncertainties in evaluating the accident progression and the fuel debris distribution and characteristics. To reduce such uncertainties, a specimen simulating the residual heattype reactor core and lower support structure was heated up with a plasma torch and the decay, melt, and migrating behavior of the core substance was observed.

30 cm

[FY2017 Research Details]

The specimen was heated up simulating the residual heat system up to the assumed temperature in the lower core area at the time the core substance in Unit 2 slumped (migrated to the lower plenum) and the results were evaluated.

[Final Results]

The following knowledge was obtained and provided it in the estimate diagram of fuel debris distribution.

- [1] High-temperature core fuels form partial blockage; however, residue fuel columns tend not to fuse with each other and presumably have macroscopic permeability against the gas phase (and the liquid phase) including when they have decayed (similar to the loose core debris in TMI-2).
- [2] High-temperature fuels would maintain their column shape up to nearly the melting point, and they hardly move in the form of a pellet. If fuels in units decay due to a load, etc., the decayed high-temperature fuel would stay in the reactor core (with no change to the axial position), presumably allowing only part of the high-temperature fuels to enter the support structure in the process leading to the core melting.

Reflection in the estimate diagram of fuel debris distribution

- >Combined with the knowledge obtained in the "event sequence analysis at the time of core material slumping," the results were used to estimate the core sub-structure fracture mode in Unit 2 and the properties (mainly solid) of the fuels that still exist in the lower plenum.
- >The knowledge that would enable more accurate evaluation of the RPV fracture mechanism, the fracture mode, the fuel debris properties when the RPV was damaged, and the outflow property.



Two planes of channel boxes (Zr) Simulated fuel rod $(ZrO_2) \times 48$ pcs

CMMR4

Control rod blade

Improved the system so it would allow easier heating. *No change to the max output of the plasma heatup.

Heated up the upper part to 2.600 K or higher. Fuel top



Observation angle of the image below

Partial blockage

A small molten pool formed due to the melt of oxides (the lower power blocked due to solidification) Space in which the control rod blade melted down deposits and (partial) blockage formed by molten metal (Fe or Zr) including boron

> Fuel rods do not decay if exposed to high temperature



E



2.b) Evaluation of Chemical Characteristics of FPs

Purpose

To decommission the Fukushima Daiichi Units 1 to 3, it is essential to identify the distribution and chemical properties of cesium accounting for a major part of the gamma source. Under this project, apart from the typical chemical species in the evaporation, migration, and condensation processes (CsI: cesium iodide, and CsOH: cesium hydroxide), a study on the possible presence of compounds that are believed to be relevant to evaluating Fukushima Daiichi (molybdate compound, silicate compound, boronic acid compound, etc.) and their impact levels is conducted. If there is uneven distribution of cesium with poor solubility --- a factor that would negatively influence the top-entry method --- in the upper reactor core region due to the reaction with the upper-structure material (steel), was also tried to be found out. Furthermore, a study was conducted on the mechanism by which insoluble cesium particles/powder dust are generated and their migration routes.





(2)-[2]-1 Reaction with Cesium and Steel Materials, and Re-evaporation

[Overview and Objectives]

- Overview: Conduct a test on Cs adsorption in steel materials and its re-evaporation, using parameters such as temperature and atmosphere to enhance knowledge of the Cs chemisorption behavior, thereby developing a Cs adsorption model that takes major compounds generated by adsorption into account.
- Objectives: Verify the existing model through a replication experiment on Cs adsorption and evaluate the chemical and thermodynamic properties of relevant compounds to develop a model that takes various chemical conditions, such as re-evaporation and Cs concentration variation, into account.

[FY2017 Research Details]

- (i) Evaluation test on adsorption and re-evaporation behavior An experiment on Cs absorption was performed, using parameters such as temperature and atmosphere, and the effects of various chemical conditions was evaluated.
- (ii) Development of an adsorption and re-evaporation model The results of the Cs adsorption experiment based on the mass transfer theory and thermodynamic data on Cs adsorption products were analyzed and modeling to consider the effects of chemical conditions such as the CsOH concentration in the gas phase was performed.

[Final Results]

- The influence of chemical conditions were experimentally clarified, such as CsOH concentration in the gas phase, atmosphere, Si concentration in steel and temperature, on the Cs adsorption volume. A chemisorption model based on the test values was developed.
- By using the model formula, the sensitivity (fluctuation range) of Cs adsorption volume to the dryer and separator according to changes in chemical conditions, such as CsOH concentration in the gas phase, was exhibited.

$$\begin{split} & \begin{array}{c} \text{Chemisorption model developed in this study} \\ & \begin{array}{c} v_d \cong 7.027 \, \frac{\sqrt{C_B}}{C_g^{0.5225}} \exp\left(-\frac{109000}{2RT}\right) \\ \text{(*): The adsorption volume was calculated using: } \frac{1}{A} \frac{dM_d}{dt} = C_g v_d \\ & \begin{array}{c} v_d: \text{ reaction rate constant [cm/s]} \\ & C_g: \text{ CsOH concentration in the gas} \\ & \text{phase [}\mu\text{g/cc]} \\ & C_B: \text{ Si concentration in steel [wt.%]} \\ \end{array} \end{split}$$



Figure 1. (Estimate) Cs adsorption volume to the dryer/separator in 1F2 where structural material temperature and CsOH concentration in the gas phase were constant for 3 hours

(2)-[2]-2 Evaluation of Particulate Cesium Compounds (1. Test on Reaction and Solidification in the Gas Phase)

[Overview and Objectives]

To clarify the generation mechanism of insoluble cesium particles, a test on reaction and solidification in the gas phase in FY2017 was conducted by using (1) Mo, (2) Zn coating, and (3) Ca as evaporation sources in addition to the standard simulation conditions of Cs-Si-O that were examined in FY2016.

[FY2017 Research Details]

An additional test was performed to examine the following possibilities:

- [1] Generating insoluble Cs particles in the environment where molybdate coexists.
- [2] Entry of the Zn coating component used for the S/C inner wall into insoluble Cs particles.
- [3] An insulation material or concrete are the original component of insoluble Cs particles.

[Final Results]

- [1] When the evaporation source, Mo, was added to the simulation test conditions, soluble Mo-Cs-O particles were produced, but no insoluble cesium particles were produced (Right figure (a)). In the molybdate-bearing environment, it is believed that no insoluble Cs particles are produced; presumably they were produced in the early phase of core damage (where FP composition was Cs>>Mo).
- [2] When the coating fragments of the S/C inner wall were added as an evaporation source, insoluble cesium particles came to contain Zn depending on the coating fragments' loading volumes (Right figure (b)).
- [3] When Ca-bearing material was added as an evaporation source, insoluble cesium particles came to contain Ca depending on the evaporation property of the Ca-bearing material (Right figure (c)). Presumably, neither concrete nor insulation was involved in generating insoluble Cs particles.







(b) Zn-bearing cesium particles (Si: 56%, Cs: 17%, Zn: 22%-metallic element)

(c) Ca-bearing cesium particles (Si: 69%, Cs: 29%, Ca: 1%-metallic element)



(2)-[2]-2 Evaluation of Particulate Cesium Compounds (2. Study on the Microstructure)

[Overview and Objectives]

According to a report, a structure having nano-sized, network-like crystalline $ZnFe_2O_4$ was found in some insoluble Cs particles. The generation mechanism of those particles was studied by simulating and evaluating the microstructure of insoluble Cs particles.

[FY2017 Research Details]A simulated sample was prepared with the following two techniques to observe the microstructure.

- [1] The sample was adjusted so it had the same composition as the insoluble Cs particles. It was loaded in a crucible and melted at 1,500°C in the air atmosphere, then quickly cooled it down with water.
- [2] SiO₂-Fe₂O₃-ZnO was adjusted so it had a composition similar to that of the insoluble Cs particles and hence had the possibility of experiencing spinodal decomposition. Then, it was melted by gas levitation and laser heating and then solidified.



Figure 1: TEM image of sample 1 (Phase identification based on electron diffraction results)



Figure 2: SEM image of sample 2 (Elementary analysis with EDS)

[Final Results]

- ✓ In the SiO₂ glass matrix of Sample [1], the generation of nano-sized crystalline ZnFe₂O₄ (Figure 1) was observed. The particle observed by Yamaguchi et al.^{*1} (a structure formed through the dynamic dissolution of ZnFe₂O₄) requires rapid cooling after it is generated at a high temperature. It suggests that this type of structure was generated at a high temperature in the RPV and then rapidly cooled in the S/C.
- Presumably, the network structure of the Zn-Fe-rich phase shown in Sample [2] (Figure 2) was generated by spinodal decomposition. The microstructure observed by Furuki et al.,^{*2} (a network crystalline structure of ZnFe₂O₄) is nano-sized and has a different dimension from the spinodal decomposition structure formed in the liquid phase mocked up under the conditions of Sample [2]. The origin of the nanostructure remains unknown.

*1: N. Yamaguchi et al., Sci. Rep. DOI: 10.1038/srep20548, 2010 *2: G. Furuki et al., Sci. Rep. DOI: 10.1038/srep42731, 2017

(2)-[2]-2 Evaluation of Particulate Cesium Compounds (3. Evaluation of Inorganic Zinc Coating)

[Overview and Objectives]

A study on the possibility that inorganic Zn coating in the S/C inner wall was the material origin of the constituent elements (Zn and Si) of insoluble Cs particles, was conducted. A simulated coating sample was prepared by referring to construction specifications and the coating components' leaching behavior was examined under the test conditions simulating the accident (with purified water at 140°C for 30 hours).

[FY2017 Research Details]

 By using ICP and IC, the coating components before and after the leaching test (Table 1) was measured. Apart from Zn and Si, a small amount of K and CI was detected.

[Results]

- Inorganic Zn coating components can only be involved with Unit 2, where the RCIC water source was switched from the CST to the S/C. Hence, the RPV of Unit 2 is the site where the insoluble Cs particles occurred in the environment.
- ✓ From the amount of K₂O supplied from the coating (on the order of 100 g) and K₂O concentration in insoluble particles (approx. 1 wt%), the amount of insoluble Cs particles generated is estimated to be on the order of more than 10 kg (Table 2).
- ✓ As the ratio of SiO₂/K₂O supplied from the coating shows SiO₂ was insufficient, the evaporation of silica from the steel materials may have contributed to the amount.

Table 2. The estimated amount of insoluble Cs particles generated (based on the potassium amount)

Items	Evaluation values	Evaluation methods
1. Zinc coating weight	2,600 kg	Coating area $(8,566 \text{ m}^2) \times \text{Ratio of the smooth}$ area $(0.8) \times \text{Coating thickness (100 } \mu\text{m}) \times \text{Coating}$ density (3.8 g/cm ²)
2. K content	3.12 kgK (3.76 kgK ₂ O)	2,600 kg \times 0.0012 (Table 1 average)
3. Amount of K ₂ O dissolved	0.752 kgK ₂ O	3.76 kgK ₂ O \times 0.2 (dissolution rate; Table 1 average)
4. Brought into the RPV	0.125 kgK₂O	Dissolution amount \times Amount of water evaporated/Total water amount (500 m ³ /3,000 m ³)
5. Amount of Cs particles generated	12.5 kg	0.125 kgK ₂ O/0.01 (K ₂ O content by Kogure et al.)

Table 1. Chemical analysis results of the coating before and after the test

				IC	P			IC	
No.		Zn	Si	к	Mg	Ti	AI	CI	Balance
					ma	ss%			
	Before test	79	4.3	0.13	0.03	0.06	1.4	0.17	14.91
1	After test	74	3.8	0.11	0.01	0.05	1.4	0.012	20.62
2	Before test	78	4.0	0.16	0.03	0.07	1.6	0.13	16.01
	After test	74	3.7	0.12	0.01	0.05	1.4	0.010	20.71



(2)-[2]-2 Evaluation of Particulate Cesium Compounds (4. Oxidation and Evaporation Behavior of Silicon in Steel Material)

[Overview and Objectives]

A study on the oxidation behavior of stainless steel and the existing state of silicon in such oxides under the temperature range and atmosphere containing hydrogen and water vapor that presumably simulate the temperature range and atmosphere at the time of the Fukushima Daiichi accident, was conducted. The thermodynamic properties of Cs-Si-(Fe)-O as a basic structure of cesium compounds are clarified.

[FY2017 Research Details]

- As shown in Figures 1 and 2, the Si distribution state in metallic phase near the metallic phase obtained by heating up SUS304 grade stainless steel was analyzed, in which Si content changed under water vapor atmosphere.
- As shown in Figure 3, using a thermodynamic database, the possibility that hydroxide and suboxide are the silicon sources of the aforementioned particles was estimated and studied.
- A Cs₂O-SiO₂ melt and copper, as basic structures, in a graphite crucible under coexistence of CO was equilibrated and the SiO₂ activity determined.

[Final Results]

IRIDIZE

- The existing state of Si oxides is divided mainly into Fe_2SiO_4 and SiO_2 , largely depending on the state of the oxide layer of steel material.
- When the oxidation rate is high, the oxide layer is separated into two, and Si exists in the inner oxide layer as Fe₂SiO₄; then, it is transformed to SiO₂ as the oxide layer peeled off due to long-time heating.
- Thus light was shed on the atmospheric dependence of silicon source in insoluble cesium particles and the dependence of SiO₂ activity in cesium silicate melts on Raoult's law.



Figure 1. Si distribution in stainless steel containing 1% Si, which was oxidized for 60 minutes at 1,200°C under atmosphere of H_2O/Ar

Figure 2. Si distribution in stainless steel containing 1% Si, which was oxidized for 60 minutes at 1,200°C under atmosphere of $H_2/H_2O/Ar$



Figure 3. H_2/H_2O ratio dependence of vapor pressure of SiO and Si(OH)₄

(2)-[2]-2 Evaluation of Particulate Cesium Compounds (5. Relation with the Accident Progression Sequence)

Time sequence	Generation process
During normal operation	 The major portion of Cs stays in the pellets, but some portion precipitated at the grain boundary. Mo, as intermetallic compound, precipitated mainly in crystal grains.
Earthquake (14:46, N	lar 11), Tsunami, Loss of AC power (15:40–15:41, Mar 11), Switching of RCIC water source (CST to S/C)
While RCIC is operating	 S/C water temperature rose → Exceeded the heat-resistant temperature of the coating → Coating components leached → Coating components migrated due to the RCIC water flow → Evaporated to dryness (Si, Zn, K) on the boiling surface in the RPV
Stop of RCIC (around	9 am, Mar 14), Forced depressurization of RPV (18:00, Mar 14), Decrease in water level, Start of core heating
Core heating	 Fuel cladding tube failure Evaporation of Fe crud and coating components (Zn, Si, and K) attached to the surface of fuel cladding Evaporation of Zircaloy component (Sn) Meltdown of control rods (Meltdown forming Fe-B eutectic, without B's involvement) Fuel heating during the meltdown of the upper tie plate, oxidation and evaporation of Si Discharge of Cs due to grain boundary connectivity of pellets (>1,500°C) Liquid phase of Cs₂SiO₄-SiO₂ system → Densification and spheroidization of the condensation phase
Discharge from RPV	 As a result of opening the safety valve relief (21:20, Mar 14), spherical cesium-bearing particles were rapidly discharged from the RPV, along with a large amount of water vapor and non-condensable gas → Rapid cooldown→ Suppressed nucleation and spherical precipitation of ZnFe₂O₄ → Generation of insoluble Cs particles Most of the particles were captured in the S/C water. Some of them were discharged to the environment.
Observed at environm	nental monitoring facilities in the Tokyo metropolitan area, including Meteorological Research Institute (7:00–16:00,





IRID

©International Research Institute for Nuclear Decommissioning

(2)-[2]-3 Optimization of the Evaluation Model for Cesium Compounds

[Overview and Objectives]

- Overview: Optimize the adsorption model regarding the products of reaction between CsOH and SUS304 steel under an oxidized/reducing atmosphere based on the JAEA experiment results, and develop a generation model of particulate cesium.
- Objectives: Introduce the adsorption model and the silicon generation model into SAMPSON, and develop a chemical characterization tool, based on the analysis results of the amount of silicon generated.

[FY2017 Research Details]

(i) Optimization of the adsorption model

A correlation equation for the temperature-dependent adsorption rate was used in a reducing atmosphere based on the existing data and it was determined that the effect of CsOH concentration can be evaluated using the corrected formula based on this project's results (2)-[2]-1.

(ii) Development of a generation model of particulate cesium It was determined that the Si discharge sources were the evaporated Si component in SUS core structure materials, such as the control rod blade, core support plate, and the leached Si component in the S/C inner wall coating. Then, an Si discharge model and a particulate cesium generation model was developed.

[Final Results]

IRIDIAE

- The optimized adsorption model and the particulate Cs generation model in SAMPSON was incorporated.
- A tool to evaluate the validity of the branching ratio "f" was developed based on the study results of this project (2)-[2]-2.



50

Figure 1. Temperature dependence of adsorption rate



Figure 2. Generation model of particulate cesium

(2)-[2]-4 Analysis and Evaluation of the Actual Units Using the Upgraded Model

[Overview and Objectives]

Overview: Conduct analysis of Units 1 to 3 using SAMPSON, study the consistency with items such as actual measurement data that can affect the atmosphere, and evaluate the reliability of analysis results Objectives: Evaluate the cesium distribution in the RPV and PCV based on the SAMPSON analysis results

[FY2017 Research Details]

- (i) Analysis under the conditions of Fukushima Daiichi plant The Units 1 to 3 using the SAMPSON were analyzed, for which the improved Cs behavior model was incorporated.
- (ii) Study on the factors that affect Cs behavior
 - Cs adsorption mainly occurs in an oxidized atmosphere, and adsorption in a reducing atmosphere (P(H₂) > P(H₂O)) lasted for a short time.
 - Generating particulate Cs needs a greater amount of Si than that coming from the core structure materials, requiring Si from the coatings inside the S/C. Therefore, the study was conducted only on Fukushima Daiichi Unit 2, where RCIC was run by using the S/C as the water source.

0.3 200 Amount of Si produced from the core Amount of Cs produced from 160 the fuel 0.2 120 Si [kg] 80 0.1 40 0.0 0 79 Time[h] 78 80

Figure 1. Time dependency of the amounts of Cs and Si generated in Fukushima Unit 2

[Final Results]

- ✓ Analysis was conducted and the Cs distribution in the RPV and PCV was calculated.
- ✓ It is believed that the D/W inside of Fukushima Daiichi Unit 2 has the highest contamination level when comparing Units 1–3. Meanwhile, Unit 2 analysis results showed almost no cesium on the D/W inner wall, inside the pedestal, or on each Fukushima Unit. This result indicates that the stage of simulating the current cesium distribution in each Fukushima Daiichi unit has not been reached.

✓ Current understanding and handling of the analysis codes are insufficient for analyzing the phenomenon.

IRID

(2)-[2]-5 Analysis of Samples Collected at Fukushima Daiichi NPS

[Overview] The samples collected at Fukushima Daiichi NPS such as deposits in the contaminated water in the PCV of Unit 1, contaminant attached onto the blockage inside the TIP piping in Unit 2, and a surface of a protective covering piece on the Unit 2 operation floor were analyzed. A detailed observation by performing FE-TEM, with a focus on U-bearing particles was also conducted, and the samples' microscopic characteristics were sorted out.

[FY2017 Research Details and Results]

[2] Detailed analysis of contaminated samples from Fukushima Daiichi NPS

- The following was found in the deposits in the contaminated water in the PCV of Unit 1: rust containing Fe and Zn; C-bearing material; structure material component; glass material; Cu; Pb; Zr; and U-bearing particles containing Cu (Figure "a" in the right).
- In obstacles collected from the TIP piping at Unit 2, an area containing Zr apart from the structure material components and Mo was found.
- The following was observed on the protective sheet on Unit 2's operation floor: uranium oxide particles that do not contain Zr; and vitreous particles containing Zn, Cs, etc. (Figure "b" in the right).



 (a) U-bearing area inside the deposits in the contaminated water of PCV in Unit 1
 (TEM-EDS mapping of the element U (right) and element Zr (left))



(b) Vitreous particles and uranium oxide on the protective sheet on Unit 2's operation floor



(2)-[3] Knowledge Utilization through International Joint Research

[Overview and Objectives]

The Benchmark Study of the Accident at the Fukushima Daiichi NPS (BSAF) Project Phase-2 was established by the Organization for Economic Co-operation and Development (OECD)/the Nuclear Energy Agency (NEA). The BSAF project has been hosted by the several operating agents including the Institute of Applied Energy (IAE) which utilizes the data obtained through the project and understand the conditions inside the reactors.

[FY2017 Research Details]

In FY2017, the final year of the BSAF Project Phase-2, using major physical models, the analytical results provided by participating organizations on the accident progression behavior is compared and studied, within three weeks after the scram. Their consistency is also studied with the actual measurements and on-site investigation results. The results were shared among the BSAF participating organizations as the Phase-2 output.

[Final Report]

- In July 2017, the following meetings were held at the Kokukaikan hall in Tokyo: the PRG (Program Review Group) meeting; MB (Management Board) meeting; and the joint session with other projects for the Fukushima decommissioning. The past and latest results of the internal investigations and Japan's views regarding the accident scenario based on the knowledge obtained so far, were reported on . Opinions with experts from participating organizations were exchanged. The following were discussed: the results of the comparisons among the preliminary analyses, actual measurement data and on-site investigations concerning the accident progression, fuel debris distribution and FP distribution within the three weeks after the scram; and the structure of the final report.
- In January 2018, the PRG, MB and joint sessions at the OECD/NEA headquarters in Paris were had. The evaluation results that the participating organizations provided on the accident progression within three weeks after the accident were compared against the actual measurement data and the on-site investigation results. It was confirmed that our understanding of the accident progression significantly deepened from Phase-1, as is demonstrated, for example, by the basic consistency between the evaluation results concerning the FP discharge amount and the amount discharged to the environment. The knowledge obtained through such discussions with overseas institutions was used, for our comprehensive analysis and evaluation, which helped improve the estimation accuracy on the fuel debris distribution.



Comparative example of observation and analysis results of Cs-137 depositsion



3. Research Details and Results Summary for FY2017



3. Research Details and Results Summary for FY2017

(1) Comprehensive analysis and evaluation of the conditions inside the reactors

[1] Comprehensively analyze and evaluate based on the Units data and other R&D project results

The conditions inside the RPVs and PCVs were comprehensively analyzed and evaluated based on the results of the following initiatives: "analysis and evaluation based on on-site information;" "analysis and evaluation based on the data at the time of or after the accident and inverse problem analysis;" and "analysis and evaluation utilizing analysis codes." Then, the estimate diagrams of fuel debris distribution and FP distribution was developed.

[2] Establish a database for comprehensive analysis and evaluation To share knowledge with overseas institutions more efficiently, the English search function was added to the database and the graph display function for the measurement data, etc. collected mainly within three weeks after the accident was improved. The adding of documents and data with search tags to the database that was developed under this project, contributing to a more efficient, comprehensive analysis and evaluation was also continued.



3. Research Details and Results Summary for FY2017

(2) Estimation and evaluation of fuel debris behavior and fission product behavior and its characteristics that would be useful for the comprehensive analysis and evaluation

[1] Reduction of uncertainties through analytical techniques

Sensitivity and other analyses were performed using the accident progression analysis codes, taking the boundary conditions and analysis models into account, with regard to the events assumed to have occurred inside the reactors. Thus knowledge was obtained that will be useful for comprehensive analysis and evaluation.

[2] Evaluate FPs' chemical characteristics

When evaluating the FPs' chemical characteristics, light was shed on Cs in particular, as it was believed the information would be important for decommissioning. Cs distribution and its chemical characteristics were studied, such as: identifying chemical species requiring special consideration, apart from the typical chemical species of CsI and CsOH; the production amount of insoluble Cs particles observed in the environment; and the possibility of uneven distribution of Cs due to a reaction with the structures at the upper portion of the RPV. The samples collected on-site were also analyzed and the composition and spatial distribution of uranium and FPs to understand the conditions inside the reactors was studied.

[3] International joint research to use knowledge inside and outside Japan

By using a database that was developed for the project and through managing an international joint research project (OECD/NEA BSAF Project Phase-2), the accident progression scenario and plant-related information was shared with overseas institutions. The results provided by participating organizations concerning the accident progression, fuel debris distribution, and FP distribution in the first three weeks after the accident were also compared with the measurement data and the on-site investigation results. As a result, our shared understanding about the accident progression and plant status deepened compared to Phase 1, leading to a remarkable reduction in the variety of analysis results from participating organizations and to the basic consistency of the evaluated FP discharge amount with the measured discharge to the environment. This deeper understanding of the accident progression contributed to improved accuracy of fuel debris distribution estimates. The knowledge obtained through the discussions with overseas institutions was also used for our comprehensive analysis and evaluation.



Attachment

Attachment 1. Project Organization Chart (detailed)



©International Research Institute for Nuclear Decommissioning

Attachment 1. Project Organization Chart (detailed)



International Research Institute for Nuc	lear Decommissioning (IRID)	(Consignment research)	
O Pr (1)-[1	esearch management of implementation plans, their progress status, etc. roject results summary and information dissemination 1]-1 Extraction of issues for further research to reduce uncertainties of the estimated conditions inside the reactors	The University of Tokyo (outsourcing)	(2)-[1]-2 Inverse problem evaluation using virtual reactors
	1/2 Comprehensive analysis and evaluation based on the data from the actual units and other research lopment results	Hitachi Power Solutions (Consignment research)	(2)-[1]- 2 Inverse problem evaluation using virtual reactors (Analysis and evaluation of the cooling characteristics of debris considering water injection)
(1)-[1]-1 Extraction of issues for further research to reduce uncertainties of the estimated conditions inside the reactors 1]-2 Comprehensive analysis and evaluation based on the data from the actual units and other research looment results	(Consignment research) Waseda University (Consignment research)	(2)-[1]-3-(a)-4 Event sequence analysis at the time of core material slumping [MELCOR analysis]
(2)-[1/3 (a) (4) Event sequence analysis at the time of core material slumping 1/6-1 Simulated fuel assembly degradation test 2] Analysis and evaluation of chemical forms of FPs	The University of Tokyo	(2)-[1]-3-(a)-4 Event sequence analysis at the time of core material slumping (Analysis of pipe deformation)
		(outsourcing) Sojitz Machinery Corporation	(2)-[1]-6-1 Simulated fuel assembly degradation test (plasma heatup experiment)
)1 Extraction of issues for further research to reduce uncertainties of the estimated conditions inside the reactors 2 Comprehensive analysis and evaluation based on the data from actual units and other research developments results	(outsourcing) Inspection Development Company Ltd.	(2)-[1]-6-1 Simulated fuel assembly degradation test (Service contract for CT imaging in the plasma melting test)
(2)-[1	 F Evaluation of the concrete ension state at the pedestal using sensitivity analysis Facular progression analysis using the analysis code 	(Consignment research) Osaka University	(2)-[2]-2 Evaluation of particulate cesium compounds (Property evaluation test for cesium-bearing glass)
		(Consignment research) Tokyo Institute of Technology	(2)-[2]-2 Evaluation of particulate cesium compounds (Study for evaluation of thermodynamic quantities of Cs-Si-
Systems & Solutions (1)- [1	11 Extraction of issues for further research to reduce uncertainties about the estimated conditions inside the reactors 1/2 Comprehensive analysis and evaluation based on the data from the actual units and other research development results 1/3 Accident progression analysis using the analysis code	(Consignment research) Tohoku University	(Fe)-O system compound) (2)-[2]-2 Evaluation of particulate cesium compounds (Study of silicon behavior in steel materials)
(.)	(outsourcing)	(outsourcing) Art Kagaku Co.,Ltd.	(2)-[2]-1 Response between cesium and steel materials, and re-evaporation (Evaluation test for adsorption and re-evaporation behavior)
	Electric Power Research Institute (EPRI), USA using the analysis code	(outsourcing) Nippon Nuclear Fuel Development Co., Ltd.	(2)-[2]-2 Evaluation of particulate cesium compounds (Study on the production conditions of particulate cesium)
		(outsourcing) Nippon Nuclear Fuel Development Co., Ltd.	(2)-[2]-5 Analysis of samples collected at Fukushima Daiichi Nuclear Power Station (Analysis of 1F contamination samples)

The Institute of Applied Energy (IAE)

IRIDIAE

AE	(1)-[1] Comprehensive analysis and evaluation based on the data from the actual units and other research development results
	(1)-[2] Establishment of a database for comprehensive analysis and evaluation
	(2)-[1]-3 Accident progression analysis using the analysis code
	(2)-[2] Analysis and evaluation of chemical forms of FPs
	(2)-[3] International joint research to make use of knowledge inside and outside Japan
	(outsourcing)
	Mizuho Information & Research Institute, Inc.
	(1)-[2] Management and maintenance of DB site and upgrade of display functions
	(2)-[2] Coding work associated with the FP model introduction
	(2)-[3] Management and maintenance of web portal for NEA BSAF project
	(outsourcing)
	Advance Soft Corporation
	(2)-[2] Coding work associated with the FP model introduction
	(2)-[2] County work associated with the FF model introduction

(Affiliated organizations) TEPCO