AN APPROACH TOWARD EVALUATION OF FP BEHAVIOR IN NPPS UNDER SEVERE ACCIDENTS

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ABSTRACT

Procedures for evaluation of fission product (FP) behavior under severe accident (SA) conditions should be established to determine radioactive contamination in the major areas in Fukushima Daiichi Nuclear Power Plant for suitable preparation for decommissioning works and, at the same time, to prepare for validation of SA evaluation codes for the BSAF Project Phase II. An approach toward evaluation of FP behavior consisting of forward evaluation and backward evaluation procedures is proposed. The forward evaluation procedures are carried out by applying mechanistic SA evaluation codes, in which FP behavior is divided into 5 categories. But there are several parameters not determined by the mechanistic models, *e.g.*, sizes and locations of defects of the primary containment vessels (PCVs) to determine accidental FP leakage rate from PCVs to the major areas. Those parameters can be determined by applying backward evaluation procedures, which consist of mass balance analysis to determine the amounts of suspending and depositing FPs by applying guess values for accidental FP leakage into the area, dose rate evaluation by applying the calculated FP amounts as radiation sources and then comparison of calculated and measured dose rates to determine the most realistic leakage rate. They can determine the leakage rates as solutions of the inverse problems. There are some of the backward evaluation procedures which have been successfully applied to estimate FP effluent to the environment and FP sources for the contaminated water. In the paper, main frames of the FP behavior evaluation procedures are introduced with the backward evaluation procedures to determine PCV leakage parameters.

KEYWORDS
severe accident, fission product, FP transport, ^{137}Cs, radioactive contamination

1. INTRODUCTION

In order to establish the decommissioning plan for Fukushima Daiichi Nuclear Power Plant (NPP) and to carry out decommissioning works properly, confirmation of radioactive contamination level due to depositing fission products (FPs) in major buildings and rooms (designated as the major areas) is one of the key issues. If the dose rate in the areas is in excess of the permissible level, suitable dose rate reduction procedures should be prepared and carried out. For this, evaluation procedures for FP behavior, *e.g.*, transfer, accumulation and release, under severe accident (SA) conditions should be established and the validation of the procedures should be recognized worldwide.

The Benchmark Study of the Accident at the Fukushima Daiichi NPP Project (BSAF: Nov 2012 –Sep 2014) [1] has been established to improve SA codes and analyze accident progression and current core status in detail for preparation of fuel debris removal, as a part of the R&D projects for the mid- to long-term response for decommissioning of the Fukushima Daiichi NPP, Units 1 to 4.

The project brought together international experts to advance the understanding of the phenomena of severe accident behavior specific to the Fukushima Daiichi NPP accident while also improving the methods and codes for modelling such behavior. A phased approach was applied in the NEA benchmark exercise. The range of analysis for this first phase for initial 6 day events included the following:

1) To conduct a full scope analyses of Fukushima Daiichi NPP Units 1 to 3 using currently available SA integral codes.
2) To use a time span for analysis of accident events of about six days from the occurrence of the earthquake (or reactor scram).
3) To analyze in full a number of key phenomena, e.g., initial transient from rated condition to core heat-up, core melt, release of FPs from fuel, behavior of core internals, core status including debris behavior, molten debris-coolant interaction in the lower plenum, reactor pressure vessel (RPV) failure, primary containment vessel (PCV) thermal-hydraulics, MCCI (molten core concrete interaction), hydrogen generation and FP transfer.

The first phase of the project to include the initial benchmark calculations runs for 6 days has been terminated successfully. Even though there were some findings on the FP behavior through the phase I analyses, there were still lots residual subjects on FP behavior, which should be analyzed for much longer period. The major purpose of the BSAF phase II is to prepare for the benchmark studies on FP behavior analysis with suitable benchmark data sets. Major analytical procedures might be SA codes as same as the Phase I. As the benchmark studies, lots FP behavior data, *e.g.*, FP concentrations in water, their deposition amounts on surfaces, dose rate due to deposing FPs and their release into the environment, should be prepared. It is understood that sets of those data are now in preparation for the Phase II.

FP behavior analysis based on SA codes, especially those based on mechanistic models, is a kind of forward evaluation. However, even if they try to analyze FP behavior based on the mechanistic models, sometime they will face at difficulties to determine some key parameters, *e.g.*, leakage rate of FPs through pressure boundary of PCVs, which might be determined accidentally but not mechanistically. If some backward evaluations support the forward evaluations, it will be much easier to prepare for those parameters. For effective promotion of the benchmark studies, some backward evaluations have been proposed to be involved in the project. Two of them, *i.e.*, prediction of environmental emergency dose [SPEEDI code [2]] and mass balance evaluation for contaminated water [3], have already been applied. The other, *i.e.*, mass balance evaluation based on dose rate data and assumed FP leakage rates, is proposed to support the forward evaluation.

In the paper, the methodology to approach toward FP behavior analysis based on suitable combination of forward evaluation and backward evaluation procedures is introduced. And how to contribute the backward evaluation to the forward evaluation is demonstrated.

1. CATEGORIZING FP BEHAVIOR UNDER SEVERE ACCIDENT

2.1. Categorizing FP Behavior

FP behavior under SA conditions are divided into 5 categories according to the location where FPs transfer, accumulate and release (**Table 1**). The FP behavior in Category 1 can be evaluated by the mechanistic SA code, e.g., SAMPSON code [4]. For Category 2, some of the key parameters, *e.g.*, leakage rates of FP through pressure boundaries of PCV, should be assumed for evaluation. For Categories 3 and 4, FP sources for the environmental release and the contaminated water should be determined as a result of the SA code but at the present time the effluent sources have been estimated by the backward evaluation. Those should be confirmed by applying the forward evaluations.

Table 1. Categorizing of FP transportation and accumulation during and after severe accident.

Category 1: Remain in RPVs and PCVs	
Category 2: Scatter into the major areas in the reactor buildings as sources of radioactive contamination	
Category 3: Release into the environment as effluent	[environmental diffusion evaluation]*
Category 4: Release as contaminated water and be storage in the site	[contaminated water evaluation]*
Category 5: Others (scatter in the plant as contaminated rubble and soil)	

* Backward evaluations have been reported, which are shown in Table 2

The forward evaluation procedures on FP behavior are divided into 7 steps (**Fig. 1**). Firstly, the core inventories of FPs were prepared as the common FP data bases in the reactors, which were obtained with the ORIGEN-2 code [5]. The forward evaluations (Steps 1-6) are carried out based on the results of the ORIGEN-2 code [6].

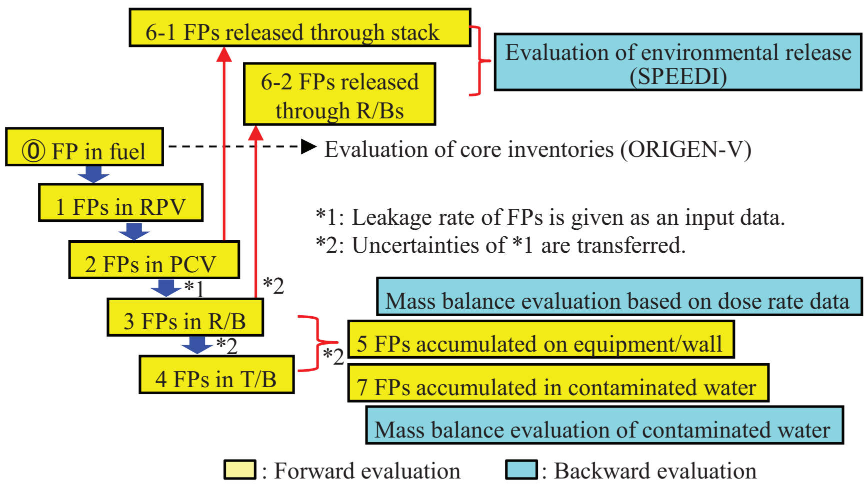


Figure 1. Evaluations of FP transfer around the R/Bs.

The steps of the forward evaluations are listed in **Table 2**, where the relationships of the forward and backward evaluation procedures are also shown.

Table 2. Major steps for evaluation of FP transportation and accumulation.

Step	Forward evaluation	Backward evaluation
0	Evaluation of total FP inventories in the cores [ORIGEN code]	
1	Release into the RPVs [SA codes]	
2	Release into the PCVs [SA codes]	
3	Release into the R/Bs [SA codes]*	• Mass balance evaluation
4	Release into the T/Bs [SA codes]*	based on dose rate data
5	Accumulation on surfaces of equipment, wall and floor in the R/Bs [SA codes]*	and assumed FP leakage rates
6-1	Release into the environment through venting processes [SA codes]	• Prediction of environmental
6-2	Release into the environment through the R/Bs [SA codes]*	emergency dose [SPEEDI code]
7	Accumulation in the contaminated water [SA codes]*	• Mass balance evaluation
		for contaminated water

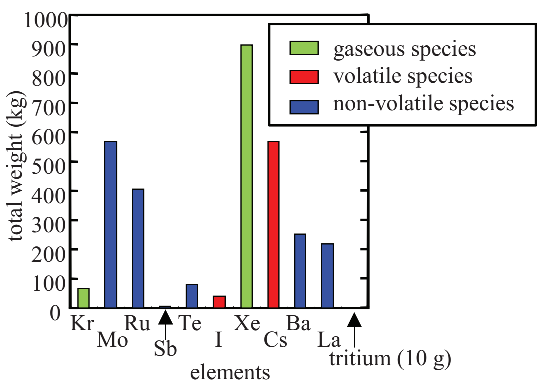
* Leakage rates from the PCV should be assumed and then confirmed by the backward evaluation

In the first two steps, all parameters can be determined mechanistically by applying suitable physical and chemical data bases as well as plant parameters. Between the steps 2 and 3, there is some gap, *e.g.*, FP leakage rates through the boundaries of PCV, which is difficult to be determined mechanistically.

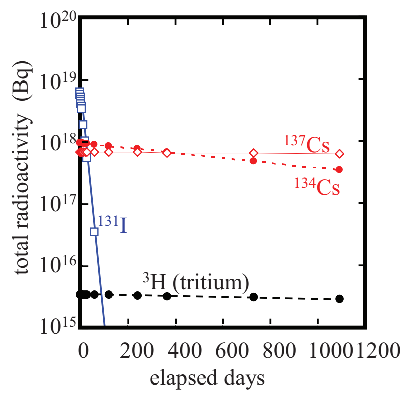
Where the leakage is and how large the leakage is might be determined probabilistically. The forward calculation can be carried out based on suitable assumption of the leakage rates. FP releases into the environment are to be evaluated based on the results of the steps 6-1 and 6-2, where much more parameters might be assumed to obtain the results. For these the backward evaluations have been carried out to determine the FP release rates based on the measured results of plenty amounts of measured data of widely distributed monitoring posts. In order to confirm the FP leakage rates through PCV boundaries, a backward evaluation is proposed, which is based on dose rate in the major areas and applied to determine surface contaminations and then the leakage rates. The details of the procedures are shown in Chapter 3.

2.3. Total Amount of FPs Generated in the Plant

The total amounts of FPs in the core can be evaluated with the ORIGEN-2 code for each of the reactors [6]. The total amount of the mass of each element for Units 1, 2, and 3 is shown in **Fig. 2** a), where FPs are divided into gaseous, volatile, and nonvolatile species. The total amount of cesium is about ten times larger than that of iodine. The dominant mass of FPs is determined by non-radioactive species. The mass of radioactive species was negligibly small. The total mass of tritium is only 10 g, which cannot be shown on the bar graph. The amounts of radioactive species decrease with elapsed time due to radioactive decay. The elapsed time-dependent radioactivity of each radionuclide is shown in Fig. 2 b).



a) Initial weight of major fission products



b) Radioactivity of major fission products

Figure 2. Major elements and radioisotopes of fission products.

3. BACKWARD EVALUATION

3.1. Previously Reported Backward Evaluation Procedures

There are two examples for the backward evaluation procedures, *i.e.,* the prediction of environmental emergency dose based on SPEEDI code [2] and the mass balance evaluation for the contaminated water [3]. The details of the procedures were shown in the references. The uncertainty in evaluation of environmental effluent by SPEEDI was also discussed in the reference, where the authors estimated a factor of 5 of uncertainty in their evaluation. As a result of evaluation of FP effluent, it was confirmed that about 1% of total amounts of radioactive cesium and iodine released into the environment [1]. The contribution of each unit could not be identified by the analysis.

As a result of the delay in restoration of the PCVs, cooling water injected into the RPVs of the Units 1 through 3 is still being recirculated through large areas from the RPVs to the T/Bs, and 85,000 tons of recirculated water should be cleaned up by removing radioactivity and chloride ions [3]. Additionally, underground water leaking into the recirculation water has resulted in an increasing amount of contaminated water stored at the site. The total amount of the contaminated water treated with the clean-up system had reached 640,000 tons by April 2015 [7, 8]. As a result of the mass balance analysis for FPs in the contaminated water, it was concluded that about 50% of the initial cesium inventory has been stored in the storage tanks with cesium adsorbent and ion exchange resin.

3.2. Mass Balance Evaluation Based on Dose Rate Data

Schematic flow diagram of FP transfer in major areas is shown in **Fig. 3**. A backward evaluation procedure based on the data base of measured radiation dose rates consists of three processes, i.e., Process 1; Mass balance analysis to determine the amounts of suspending and depositing FPs by applying guess values for accidental FP leakage into the area, Process 2; Dose rate evaluation by applying the calculated FP amounts as radiation sources and then Process 3; Comparison of calculated and measured dose rates to determine the most realistic leakage rate. Dose rate in PCV have been measured by containment atmosphere monitoring system (CAMS) [9]. Some of the measured results have been reported. There were a lot of dose rate data collected for occupational exposure control, which were opened recently [10].

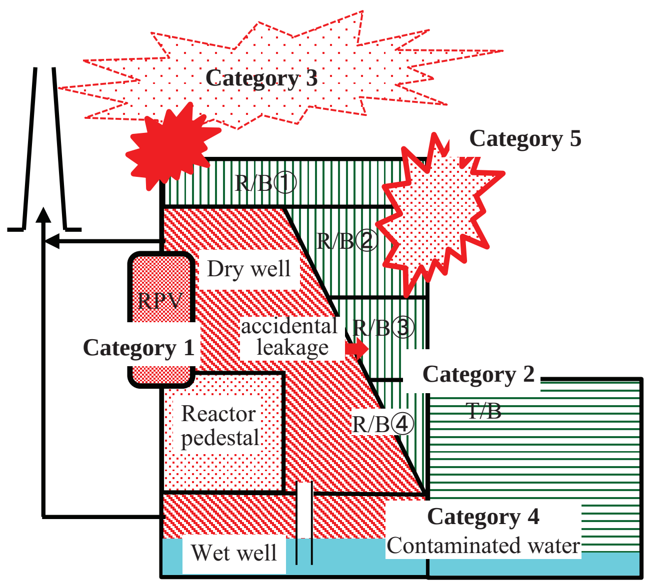


Figure 3. Major categories for FP behaviors around the reactor.

3.2.1. Process 1; Mass balance analysis

Basic equations are shown as follows. The major symbols are listed in the Nomenclature.

Concentration in atmosphere

$$dC_{RB}/dtV_{RB}=C_{RB}^{in}G_{RB}^{in}-C_{RB}G_{RB}-\delta_{RB}C_{RB}S_{RB}+\zeta_{RB}\Gamma_{RB}\Sigma_{RB}-\chi_{RB}C_{RB}V_{RB}+\chi_{RB}^{*}C_{RB}^{*}V_{RB}-\lambda C_{RB}V_{RB} \quad (1)$$

Deposition on surface,

$$d\Gamma_{RB}/dt\Sigma_{RB}= \delta_{RB}C_{RB}\Sigma_{RB}-\zeta_{RB}\Gamma_{RB}\Sigma_{RB}-\lambda\Gamma_{RB}\Sigma_{RB} \quad (2)$$

For the calculation, firstly major information on the area, *e.g.*, inner volume, surfaces of floor, walls, ceiling and major components, layout of the major components and pipes, should be collected for preparing for the input data.

As an example of the mass balance evaluation in the Process 1, where the calculated area was simplified as only room without any component, ^{137}Cs was selected as the representative FP, deposition and release rate coefficient (δ: 10^{-3} m/s and ζ: 10^{-4} s^{-1}) were obtained from analogy of radioactive crud behavior in steam and then chemical form change was neglected [11]. The amounts of ^{137}Cs suspending in the air and depositing on the floor were calculated based on Eqs. (1) and (2), and the results are shown in **Fig. 4**. The major input data are shown in Fig.4. In the case, the leakage rate was determined by defect size (diameter: 0.001 m) with choking velocity. It was also assumed that the leakage was stopped in 5 hours due to pressure suppression in the PCV.

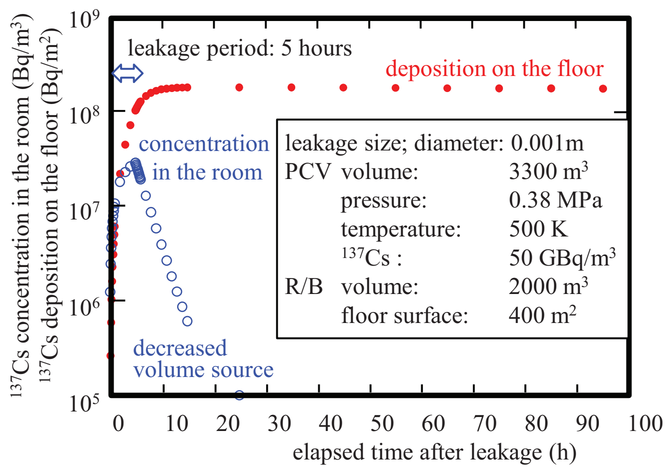
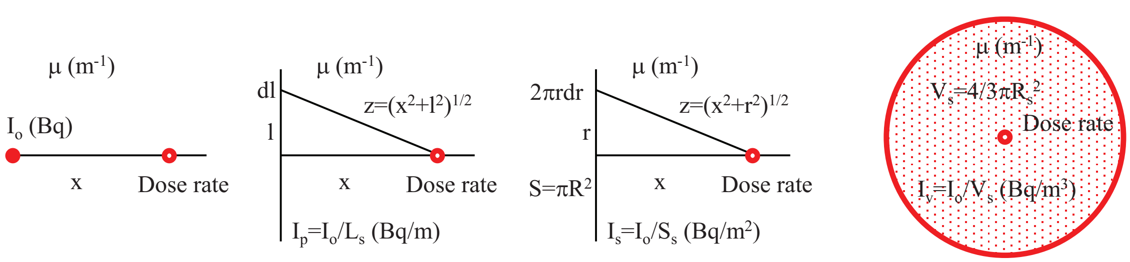


Figure 4 Results of the backward evaluation (^{137}Cs concentration and deposition).

3.2.2. Process 2; Dose rate evaluation

Radiation dose rate in the area can be determined for each of the typical source geometries, i.e., point, line, plain and volume sources. The basic formulas for the source geometries are shown as follows [12].

a) Point source (**Fig. 5** a))	$D_p = X\ I_o \exp(-\mu x)/(4\pi x^2)$		(3)
b) Line source (Fig. 5 b))	$D_l = 2\ X\ I_o/L_s/x\ F(\mu x, \pi/2)$		(4)
	$F(x, \theta) = \int_0^{\theta}\{d\theta \exp(-x/\cos\theta)\}$	F-function [13]	(5)
c) Plain source (Fig. 5 c))	$D_s = 2\pi\ X\ I_o/S_s\ E_1(\mu x)$		(6)
	$E_1(x) = \int_x^{\infty}\{\exp(-\tau)/(\tau)\}\ d\tau$	E_1-function [14]	(7)
d) Volume source (Fig. 5 d))	$D_v = 4\pi/\mu\ \ X\ I_o/V_s$		(8)



a) Point source b) Line source c) Plain source d) Volume source

Figure 5. Dose rate for typical source geometries.

Dose rate distribution for the typical source geometries are calculated in **Fig. 6**. The attenuation coefficient in the air for the calculation could be obtained from the reference [15]. The real contamination might be much more complex. Heterogeneous deposition and overlapping of dose from numerical sources are expected. Measured dose rate distribution pattern can be applied for determining source geometry. When there is no equipment and piping contributing as FP source, it is considered that the contribution of radioactive contamination on floors might be the dominant radiation source, which can be confirmed by special distribution of dose rates.

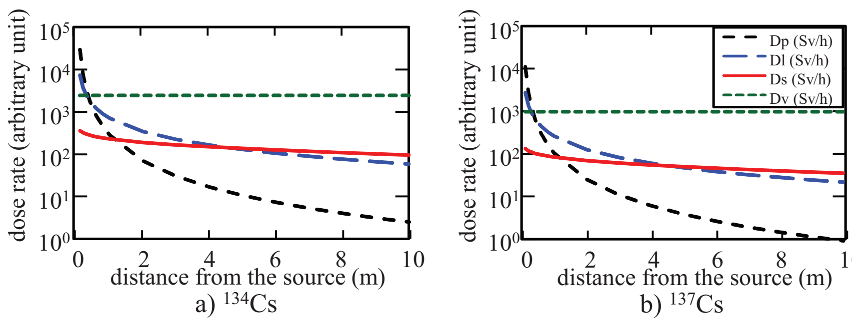


Figure 6. Dose rate for typical source geometries.

The absolute value of the dose rate can be applied to determine the contaminated FP amounts by synthesizing a combination of dose rates due to several simple sources. And then, FP effluents from the PCV to the major areas in the R/B and then leakage rates can be evaluated by analyzing the inverse problems based on the evaluated depositing amounts.

3.2.3. Process 3; Determination of leakage rate due to comparison of calculated and measured dose rates

The concentration of major FPs in PCV atmosphere in several hours after the reactor shutdown was evaluated based on SAMPSON analysis [4]. The radiation dose rate due to major FPs depositing on an infinite plane was calculated as a function of elapsed time from the reactor shutdown, which are shown in **Fig. 7**. The contribution of short-lived FPs to the dose rate was observed but it decreased rapidly and then it was controlled only by long-lived ^{134}Cs (half-life: 2.8 years) and ^{137}Cs (half-life: 30 years) with sufficient amounts of their initial inventories. After 3 years, the contribution of ^{134}Cs and ^{137}Cs to the dose rate was more than 99% and that of ^{137}Cs exceeded 50%.

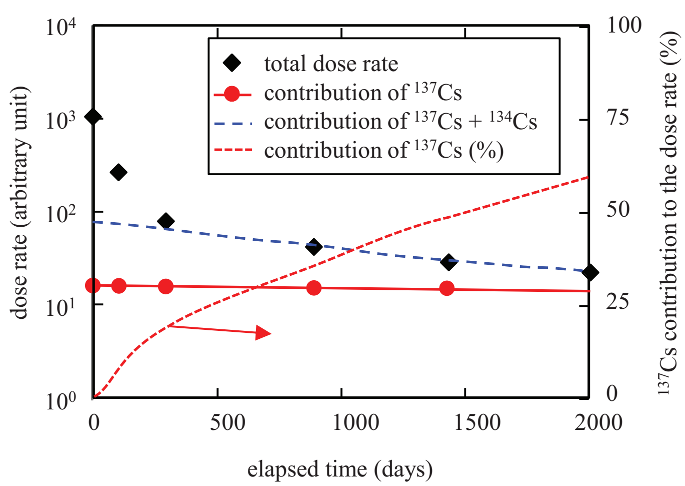


Figure 7. Dose rate from contaminated wall.

The dose rate in the area is determined by ^{137}Cs and ^{134}Cs, whose contribution to the dose rate is expressed as a function of elapsed time as shown in Fig. 7. When the dose rate was measured and the source geometry was recognized, for instance, as the plain surface, the deposing amount of ^{137}Cs on the surface can be determined by applying the relationship between dose rate and ^{137}Cs amount shown in Fig. 7.

In the example of the evaluation, dose rate from the ^{137}Cs depositing on the floor (3rd floor of R/B in the Unit 1) was selected [10]. The measured dose rates at about 30 locations were scattered within a factor of two. From three reasons, (1) long term stable dose rate, (2) lower spatial dependency and 3) no other point- and line-sources existing in the area, it could be considered that the dose rate was determined by the surface contamination, especially form the floor.

The calculated concentration and deposition amounts shown in Fig. 7 were applied to calculate dose rate by applying the attenuation coefficient in the air of 0.67 MeV gamma rays of ^{137}Cs ($9.3\text{x}10^{-3}$ m^{-1}) and the dose rate conversion factor for photon fluence ($2.7\text{x}10^{-13}$ Sv/h/(photons/m^2/s)) [15].

The concentration and deposition amounts were calculated by changing the assumed defect diameter and then the dose rate was calculated based on the obtained concentration and deposition amounts. In **Fig. 8**, the calculated dose rates are shown as a function of assumed defect diameter and they are compared with the measured ones (measured in 3 years after the accident) with some uncertainty [10]. As a result of comparison between the calculated dose rate and measured, it was concluded that the effective defect diameter might be 0.001± 0.0005 m.

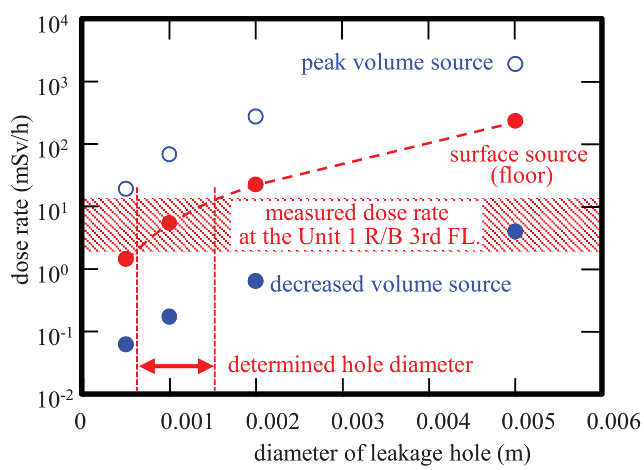


Figure 8. Results of the backward evaluation (Calculated and measured dose rates).

The calculated result is only an example to show how to apply the backward evaluation. FP transfer from one area to others, their chemical form change and their drain as contaminated water are taken into account for obtaining much more realistic evaluation. The real source geometries, e.g., more than one surface sources, line sources of piping containing contaminated water, sources due to heat exchangers contaminating on its surface and involving contaminated water and their combinations, which make the radiation fields much more complicated. Based on the dose rate data, FP leakages from the PCVs are evaluated step by step by applying the backward evaluation to multi-sources.

In the example, deposition and release rate coefficients were determined from analogy of those of radioactive crud in steam. Deposition due to gravity force is also involved in the deposition processes.

4. FORWARD EVALUATION

As an example, the results of forward evaluation, SAMPSON, are shown in **Fig. 9**. Dominant portions of ^{137}Cs are accumulated in the wet well and quite amounts of FPs are transferred in R/B but only few portions are released into the environment. The environmental release can be checked with SPEEDI analysis [2], while the amounts in the wet well, reactor pedestal and R/B can be estimated from the effluent sources for the contaminated water.

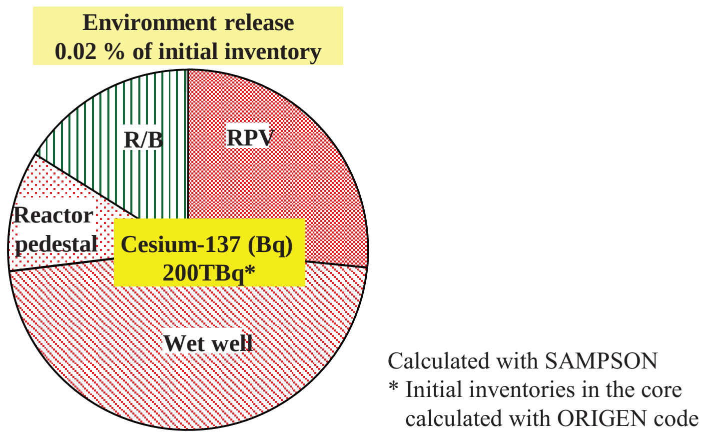


Figure 9. Calculated ^{137}Cs distribution around the No. 1 Unit reactor (100 hours after the reactor shutdown).

As a result of evaluation of the contribution of FP effluent, it was confirmed that about 1% of total FPs, radioactive cesium released into the environment. The contribution of each unit could not be identified by the analysis. But, there is still some gap between the ^{137}Cs effluent estimated with SPEEDI and SAMPSON. One of the possible reasons of the gap was from decontamination factor (DF) during wet well venting, which suggested that DF data applied in SAMPSON code should be re-evaluated.

The total amount of ^{137}Cs leakage from PCV to the R/B 3rd floor based on the dose rate analysis was much smaller than the estimated value by SAMPSON evaluation as well as contaminated water evaluation [3]. Further discussions are required to improve the evaluation accuracy. Now a tool for the backward evaluation procedure based on dose rate data has been developed. Optimization of the procedures and determination of suitable calculation constants have been started to evaluate overall FP behavior under SA.

5. CONCLUSION

The conclusions are summarized as follows;

1) Analytical procedures for FP behavior under SA conditions are going to be established to evaluate radioactive contamination in the major areas in Fukushima Daiichi NPP for suitable preparation for decommissioning works and to apply the procedures to the benchmark analysis on FP behavior based on the SA code for the BSAF Project Phase II.
2) Methodology to approach toward FP behavior analysis based on suitable combination of forward evaluation (SA code) and backward evaluation procedures was introduced
3) An approach toward FP behavior evaluation consisting of 7 step procedures was proposed to establish the targets shown above. The whole step evaluations have been carried out, in principle, by applying mechanistic severe accident evaluation codes (forward evaluation).

4) The backward evaluations support the forward evaluation procedures for determining a few parameters not determined by the mechanistic model, e.g., FP leakage rate from PCVs.
5) The backward evaluation procedures which consist of mass balance evaluation in the target areas based on sets of radiation dose rate data and determine the leakage rates as solutions of the inverse problems have been newly developed.
6) The examples of the evaluation according to three processes were demonstrated.
7) Further developments are required to apply the backward evaluation procedures for evaluating FP behavior under SA.

NOMENCLATURE

C_{cw}: concentration in the contaminated water (Bq/g) [superscript: o; initial concentration, out; in the outlet flow, u; underground water]
C_{RB}: concentration in the area in R/B (Bq/m^3) [superscript: in; in-leak, *; another chemical form]
D: radiation dose rate (Sv/h) [superscript: p; point source, l: line source, s; plain source, v; volume source]
f_{cw}: decontamination factor at the contaminated water treatment system ($f= C_{cw}/C_{cw}^{out}$)
G_{cw}: flow rate (g/s) [superscript: I; injected into the reactor, u; underground water leaking in the contaminated water]
G_{RB}: leakage rate in the area in R/B (m^3/s) [superscript: in; in-leak, out; out-leak]
I_o: source intensity (Bq) [superscript: o; total, l: line source, s; plain source, v; volume source]
L_s: length of line source (m)
S_{cw}: source amount in the reactor (Bq)
S_s: surface of plain source (m^2)
t: time (s)
V_{cw}: total inventory of the contaminated water (g)
V_{RB}: regional volume of the area in R/B (m^3)
V_s: volume of source (m^3)
x: distance from the source (m)
χ_{RB}: chemical form change rate in the area in R/B (1/s) [superscript: *; additional term from another chemical form]
X: dose rate conversion factor (Sv/h/(Bq/m^2))
δ_{RB}: deposition rate in the area in R/B (m/s)
ε_{cw}: residual portion of source a week after the accident (-)
Γ_{RB}: depositing amount in the area in R/B (Bq/m^2)
λ: decay constant (s^{-1})
μ: macroscopic attenuation coefficient in the air in the area in R/B (m^{-1})
θ: angle of elevation to the source point (-)
Σ_{RB}: surface in the area in R/B (m^2)
ζ_{cw}: release rate of species from the residual source (s^{-1})
ζ_{RB}: release rate from surface in the area in R/B (1/s)

ABBREVIATIONS

BSAF: an OECD NEA project “The Benchmark Study of the Accident at the Fukushima Daiichi NPP Project”
CAMS: containment atmosphere monitoring system
DF: decontamination factor
FP: fission product

NPP: nuclear power plant
ORIGEN: a computer code system for calculating the buildup, decay, and processing of radioactive materials
PCV: primary containment vessel
RPV: reactor pressure vessel
R/B: reactor building
SA: severe accident
SAMPSON: a severe accident analysis code
SPEEDI: a computer code system for the real-time prediction of radiation dose to the public due to an accidental release
T/B: turbine building
WSPEEDI: world-wide version of SPEEDI

ACKNOWLEDGMENTS

The authors express their sincerely thanks to Dr. Hiroshi Shirai, chief researcher of Institute of Applied Energy, for his enthusiastic contribution on SAMPSON evaluation.

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