

# Analysis for Progression of Accident at Fukushima Dai-ichi Nuclear Power Station with THALES2 code

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## ABSTRACT

The analysis of the progressions of the accidents at the Fukushima Dai-ichi nuclear power station unit 1 and 2 were performed with the THALES2 code. The KICHE code was coupled with THALES2 to simulate the chemical reaction in the aqueous phase. The analytical conditions were set on the basis of the information provided in the OECD/NEA BSAF project. The CV failures were assumed in the both cases of units 1 and 2. Addition to the DW failure case, the S/C failure case was set as the CV failure of unit 2. The transitions of melt progression and relocation of units 1 and 2 were shown as the results of the analyses. The results of the analyses have indicated that the oxidation of the zirconium of the fuel cladding tube generated approximately 1,200 kg of hydrogen in both units 1 and 2. The analytical results showed FP transition during the accidents. It is indicated that 80% of cesium and iodine distributed in the water phase of S/C due to the scrubbing. Cesium was released into the environment mainly as the form of CsOH or CsI. The release fraction has been predicted as  $10^{-3}$  for unit 1 and  $10^{-1}$  for unit 2 (both failure cases).  $I_2$  and organic iodine were formed as the result of the chemical reaction in the water phase of S/C, and their released fractions were at the order of  $10^{-3}$  for unit 1 and 2.

## KEYWORDS

THALES2, Source term, Fukushima Dai-ichi NPS accident, Melt progression

## 1. INTRODUCTION

### 1.1 background

The analysis for progression of the accident at the Fukushima Dai-ichi nuclear power station (NPS) was performed in this study. Fission products (FPs) are released from the core fuel of the nuclear power plant (NPP) in the situation of the severe accident (SA). Hence, there is the possibility of FPs leakage into the environment and the radiation exposure of public during the SA.

The Fukushima accidents occurred on March 11, 2011 as the result of the Tohoku District-off the Pacific Ocean earthquake and the following tsunami, which caused the loss of electricity and ultimate heat sink at the NPS. The accumulated heat and hydrogen generated by the metal-steam reactions increased the pressure of the reactor cooling system (RCS) and the containment vessel (CV), and then the containment venting through the suppression chamber (S/C) was operated in units 1 and 3. In addition to the operation of the containment venting, the degradation or failure of the CV of the NPS happened, resulting in the significant release of radioactive nuclides into the environment [1].

Due to their high volatility, iodine and cesium play main roles from the view point of the public exposure. The measured sampling data obtained in the situation of the accident indicates this point [2]. Therefore, the evaluation of the transportation in the RCS and CV and the environmental release of the volatile FPs especially for iodine or cesium during the accident is of importance and necessary for assessment of the radiological consequences. Additionally, the evaluation for the spatial distribution of FPs and the core debris in the NPS is valuable for technical support of the decommissioning.

The integrated analysis code THALES2 has been developed by Japan Atomic Energy Agency (JAEA) for analysis of the progression and the source term of severe accidents [3]. Analyses for source term in the early phase of the accident at unit 3 of Fukushima Dai-ichi NPS was performed in the previous studies [4 - 5]. In the analysis, THALES2 code was coupled with KICHE code, which is also developed by JAEA, to consider iodine chemical kinetics in the aqueous phase of the S/C.

## **1.2 Present study**

In the present study, the analysis has been performed with THALES2/KICHE code for the core melt progression and the FPs transportation in the accident at units 1 and 2 of the Fukushima Dai-ichi NPS under the framework of the OECD/NEA BSAF (Benchmark Study of the Accident at the Fukushima Dai-ichi Nuclear Power Station) project.

## **2. ANALYSIS CODES AND CONDITIONS**

### **2.1 Brief description of THALES2 code and KICHE code**

THALES2 code includes a large number of models for phenomena associated with core melt progression, transportation of FPs and thermal-hydraulics during severe accidents of light water reactors. Simplified models for thermal-hydraulics and core melt progression have been applied into THALES2 code so as to realize fast running capability. In the present analysis, approximately 4 through 10 hours were required for computation. On the other hand, a relatively detailed approach has been taken for FP transportation behavior in order to rationally estimate source term of important FPs. The major phenomena for FP transportation expected to occur during a severe accident, which are schematically depicted in Fig. 1, are taken into account in THALES2 code. In a computational volume, FP species are influenced by various effects such as evaporation, condensation, gravitational settling and gas-liquid partition. KICHE code plays an important role in the chemical reaction of iodine transferred into the aqueous phase.

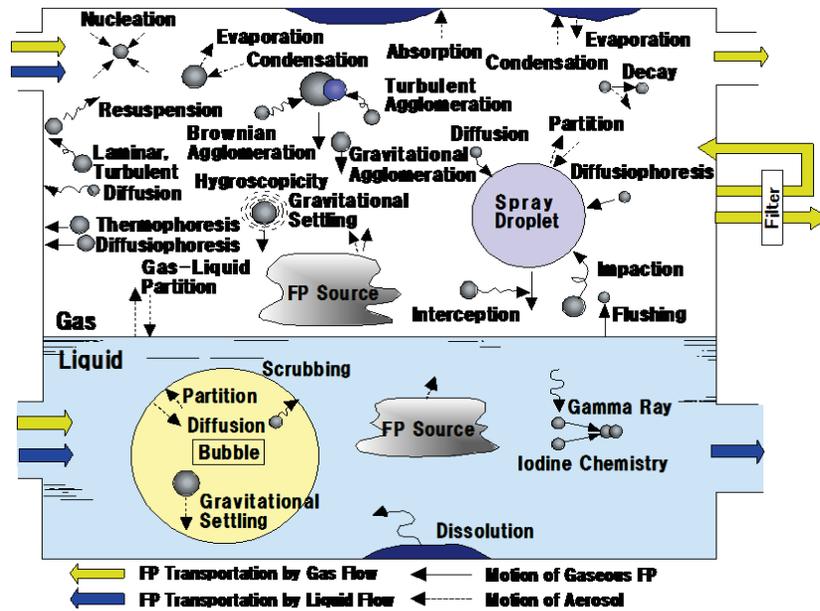


Figure 1. Major phenomena for FP transportation during severe accident modeled in THALES2 code

It is assumed in THALES2 that iodine takes a chemical form of cesium iodide (CsI) immediately after the release from the degraded fuels. Residual cesium is released as cesium hydroxide (CsOH). In KICHE code, in addition to the iodine chemistry, phenomena such as gas-liquid mass transfer and adsorption/desorption of  $I_2$  onto the structure surface are considered [6]. The brief description for coupling between THALES2 and KICHE codes is schematically illustrated in Fig. 2. In gas phase, iodine has a chemical form of CsI. Cesium iodides dissolve in water phase and disintegrate into  $Cs^+$  and  $I^-$  in KICHE calculation. Disintegrated  $I^-$  are transformed to other chemical species such as  $I_2$ , HOI and organic iodine (org-I) as the results of iodine reactions. It is noted that KICHE code is applied only into the S/C volume at present. In other words, the disintegration of CsI was not taken into account in the aqueous phases other than the S/C.

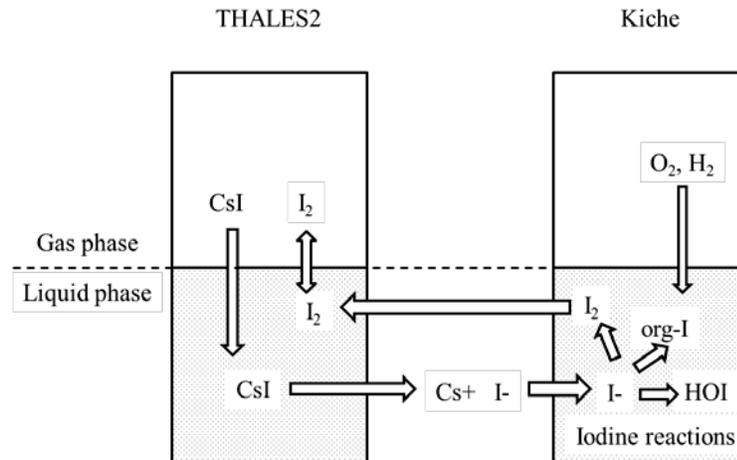


Figure 2. Iodine reactions in aqueous phase with KICHE code

## 2.2 Analysis conditions

In the present study, analyses conditions were basically set with the information provided in the BSAF project. No information concerning iodine chemistry was provided in the BSAF project since the main target of the BSAF project was to obtain technical knowledge on core melt progression. In the present analysis, the initial value of pH at 7.0 was set for the aqueous phase of the S/C and the transitions of pH for the initial condition were calculated by KICHE code. The value of the  $1.0 \times 10^{-4}$  m/s was used as the mass transfer coefficient between the gas and the liquid phases in KICHE code. The exploratory analyses were performed to determine the conditions for the flow area of the containment venting line, the failure area of the CV and the water injection rate by the fire engine pumps (0.4 and 0.2 times of BSAF project for unit 1 and 2 respectively) so as to be consistent with the measured pressure transient at the RCS and the drywell (DW). For the unit 2 analysis, it was assumed that the containment failure occurred at the DW or S/C, which was hereinafter referred as DWF or SCF case. Figure 3 shows schematics of the computational volumes and the paths of THALES2 applied in the present analyses.

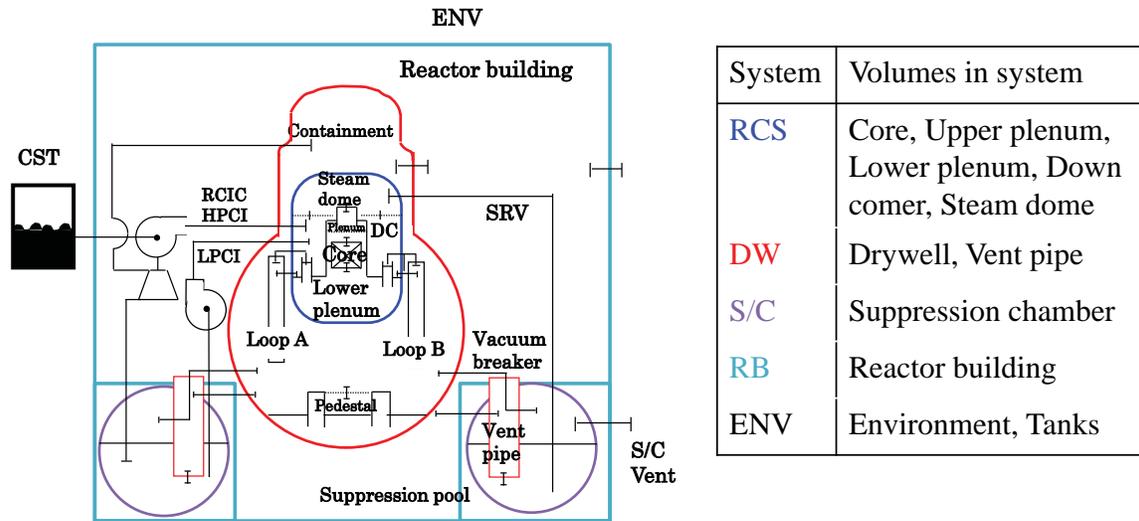


Figure 3. THALES2 computational volumes and paths

## 3. RESULTS AND DISCUSSIONS

### 3.1 Analysis for unit 1

The FP paths of unit 1 are depicted in Fig. 4. FP reaches environment through the containment venting system or the leakage of RB. The results of the analyses for the pressure transients at the RCS and the DW of unit 1 are shown in the Fig. 5. The analytical result indicates that the failures of the lower core support plate and the lower head of reactor vessel occurred approximately at 3.6 h and 5.3 h, respectively. The pressure of the RCS started decreasing by the failure of the traversing in-core probe (TIP) at the almost same time of the failure of the lower core support plate. Simultaneously the DW pressure increased, and then the DW pressure decreased at 23.4 h when the containment venting through the S/C was initiated to operate. It was assumed that the containment venting line was closed at approximately 24.3 h and the DW failed at approximately 50.0 h, resulting in increase and decrease of the DW pressure

respectively. It has been predicted, due to the model limitation, that the whole core materials slumped into the pedestal area of the DW.

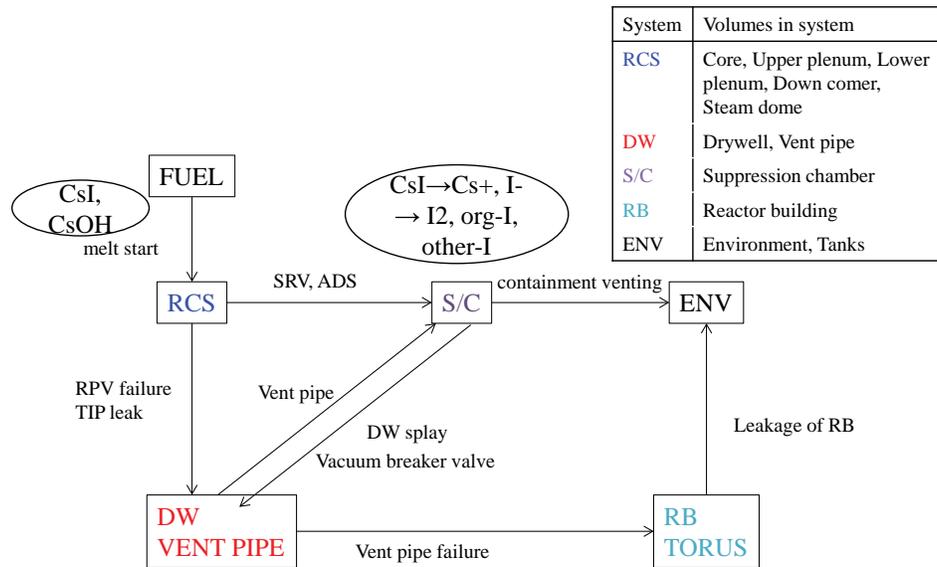


Figure 4. FP paths of unit 1

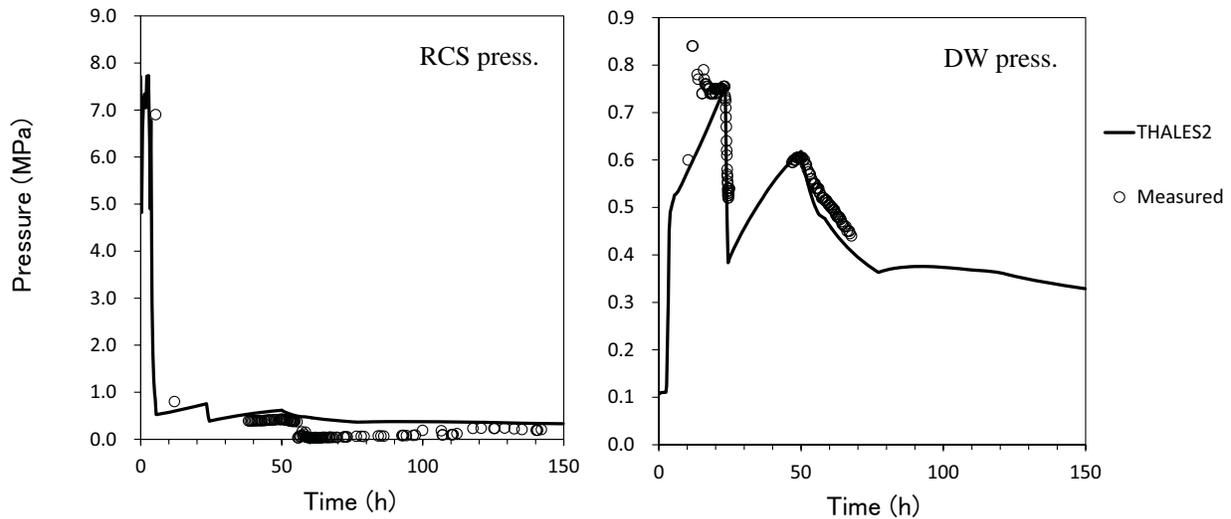
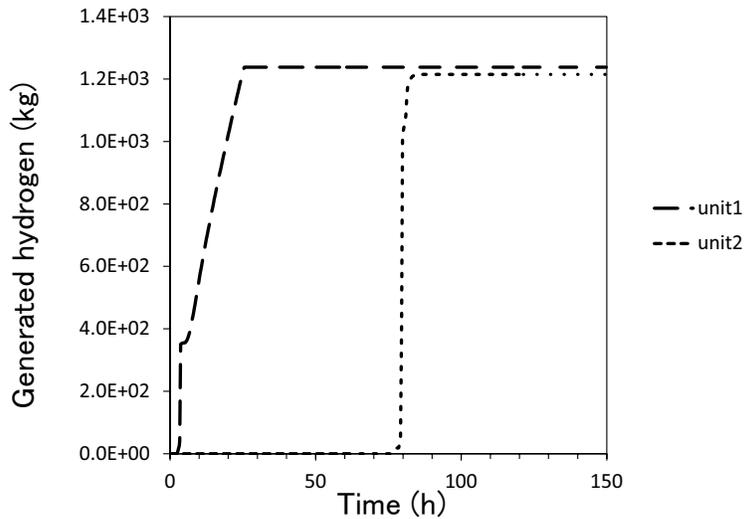


Figure 5. RCS and DW pressure transient (unit 1)

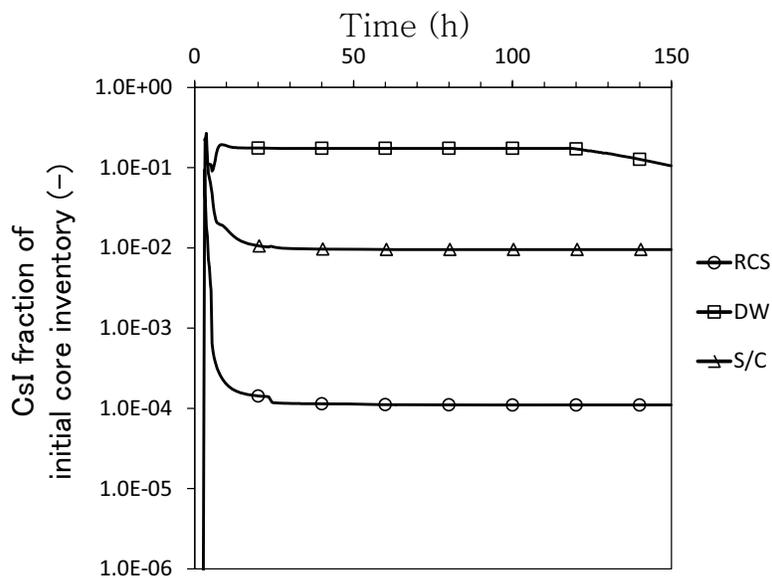
Figure 6 shows the integrated amount of the hydrogen formed by the oxidation of the zirconium composed by steam in the analyses of units 1 and 2. The amount of hydrogen was predicted to be approximately 1,200 kg in both cases, which corresponded to approximately 100 % of the initial amount of zirconium for unit 1 and 66 % for unit 2. In the case of unit 2, core materials are submerged in water after the slump into the pedestal area, at which the generation of the hydrogen stops. In the calculation, molten core-concrete interaction (MCCI) is also considered in the THALES2 code. However, it is expected that the MCCI also terminates because of the molten core submerging.



**Figure 6. Amount of the generated hydrogen**

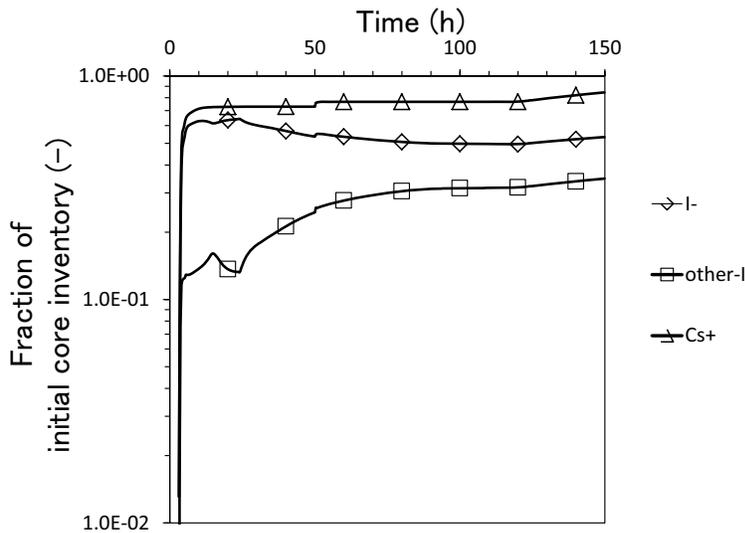
Cesium is released from the fuel into the RCS as CsI or CsOH in a physical form of gas or aerosol then they transferred to the DW or S/C. It is predicted that I, other iodine forms and Cs<sup>+</sup> were formed as the result of the scrubbing and the transformation due to the aqueous phase iodine chemistry by KICHE code.

Figure 7 shows the predicted distribution of CsI in the RCS, DW and S/C for unit 1 including the deposited amount on the structure surfaces. The vertical axis means the fraction of iodine as CsI of initial iodine inventory. Approximately 10 to 20 % of iodine of initial inventory was distributed in the DW, and there is only a small fraction of iodine at S/C or RCS. In the present analysis, the vent pipe failure was assumed to occur at 50 h. However, no clear decrease in the fraction was observed since the most CsI was deposited on the structure surfaces.



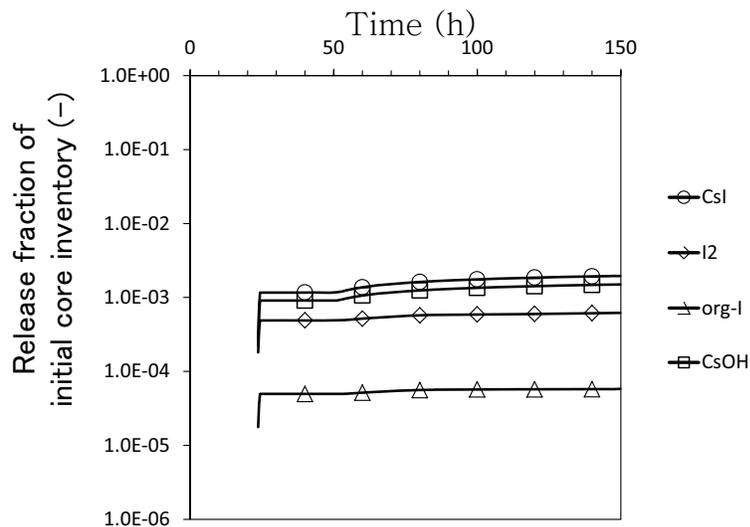
**Figure 7. CsI fraction distribution of unit 1**

Figure 8 shows the fractions  $I^-$ , other iodine (other-I) and  $Cs^+$  of the initial core inventories in the S/C. Other-I included forms of iodine other than  $I^-$  such as molecular iodine ( $I_2$ ) and organic iodine, and so on. It is indicated from the figure that most fractions (approximately 80 %) of the iodine and cesium was retained in the S/C. The variations for  $I^-$  and other-I fractions were mainly caused by chemical reactions in the water phase.



**Figure 8. Fraction of cesium and iodine in S/C water pool of unit 1**

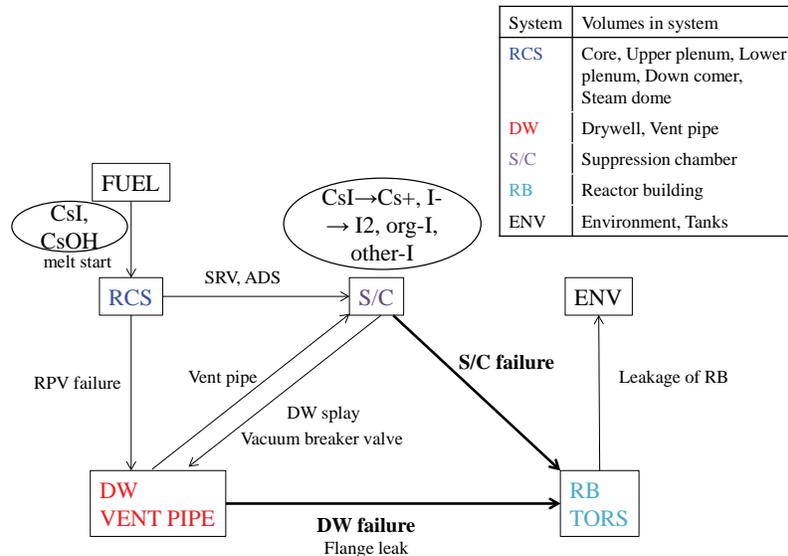
The release fraction of  $CsI$ ,  $CsOH$ ,  $I_2$  and organic iodine (org-I) into the environment is shown in the Fig. 9. The major release occurred at the containment venting around 24 h and the release rate slightly increased at vent pipe failure (50 h). It is expected that the  $CsI$  or  $CsOH$  deposits on the wall of DW or vent pipe reevaporized because of the depressurization caused by the failure. A dominant chemical form of iodine released into the environment was  $CsI$ . However, the release fraction of  $I_2$  was predicted to be comparable with that of  $CsI$  (approximately 30 % of  $CsI$  at 150 h).



**Figure 9. Release fraction to environment of unit 1**

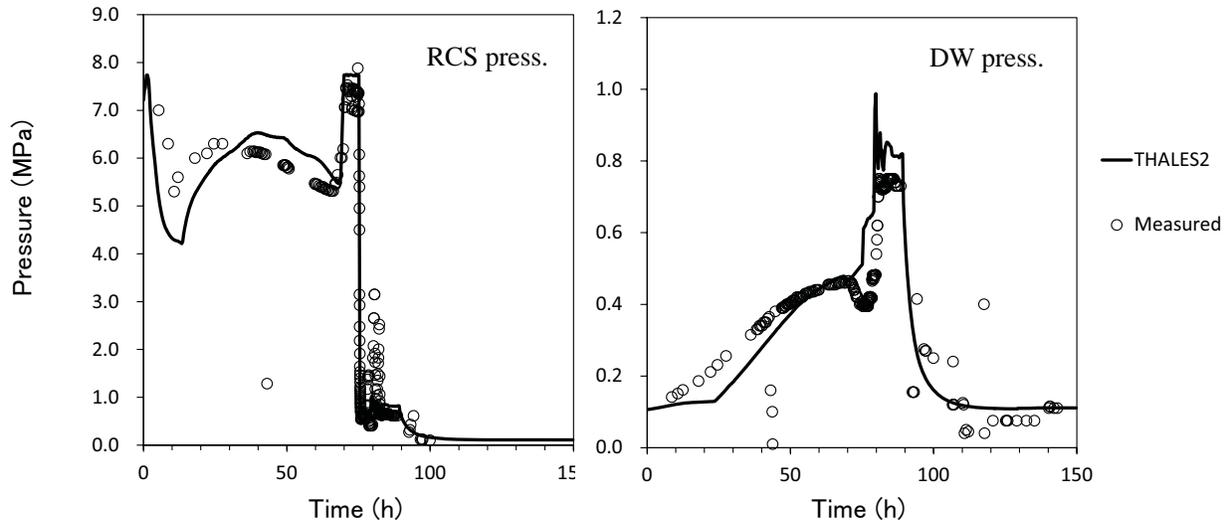
### 3.2 Analysis for unit 2

The FP paths are depicted in Fig. 10. The calculated total amount of the flow through the failure is almost ten times as much as flange leak. Therefore, it was expected that a large part of FP transferred through the failure path in the case of SCF.



**Figure 10. FP paths of unit 2**

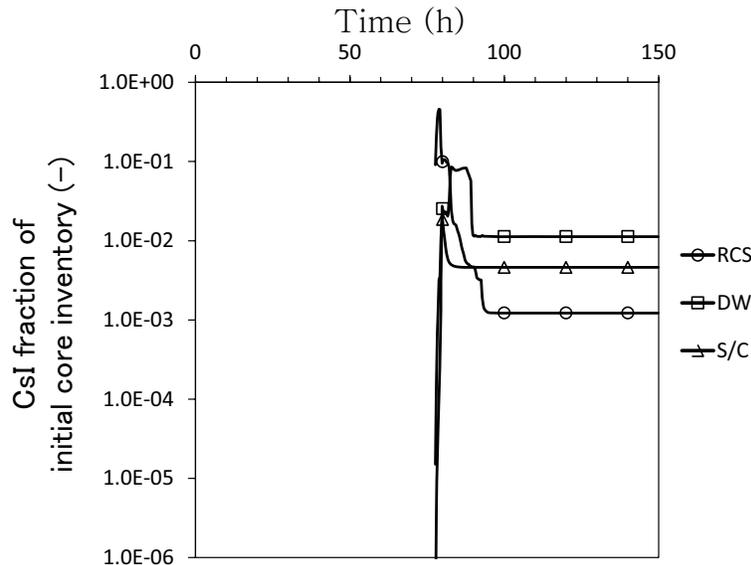
Figure 11 shows the pressure transient at RCS and DW of DWF case of unit 2 respectively. The pressure of RCS remained below approximately 6.0 MPa due to the unexpected activation of the reactor core isolation system (RCIC). The pressure of DW continuously increased until approximately 70 h because of the temperature increase of the S/C water resulting from steam inflow from the RCS through the turbine line of the RCIC system. The stop of the RCIC at 70 h caused the RCS pressure increase and then the safety relief valve (SRV) operation was conducted for the intentional depressurization at 75 h. The DW pressure started increasing at the same time up to approximately 0.8 MPa and became almost constant at this pressure. In the analysis, leakage from the DW top flange was assumed to occur when the DW pressure became higher than 0.76 MPa. At approximately 90 h the containment failure was assumed to occur in order to reproduce the sharp depressurization of the DW.



**Figure 11. RCS and DW pressure transient (unit 2)**

The core damage was predicted to start at approximately 80 h, resulting in the release of FPs from fuels into the RCS. The distribution of CsI in the gas phases of RCS, DW and S/C is shown in Fig. 12 for DWF case. The CsOH distribution was in the similar transition.

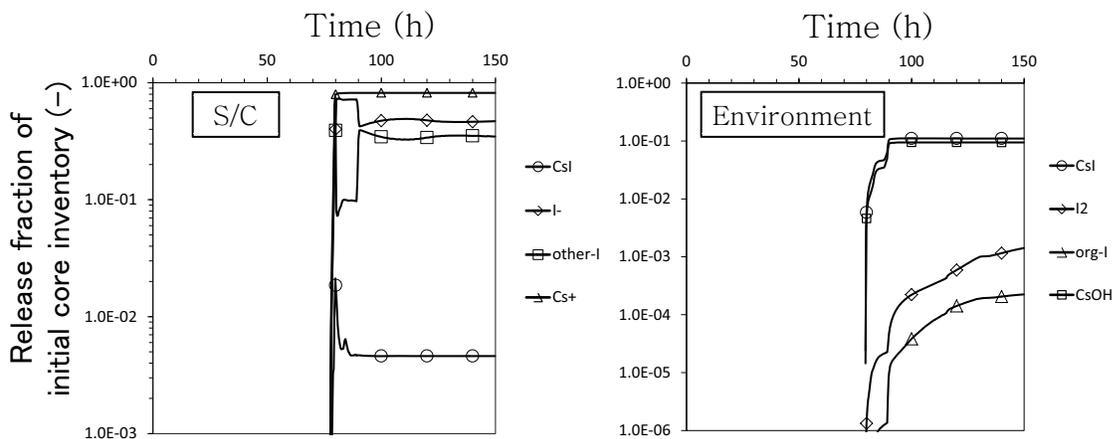
The failure of the lower head of the reactor pressure vessel occurred at 81.7 h. The CsI fraction at RCS was below the value of 10 % before the lower head failure. This means that the most of cesium and iodine transferred to the water phase of the S/C through the SRV line before the RCS failure.



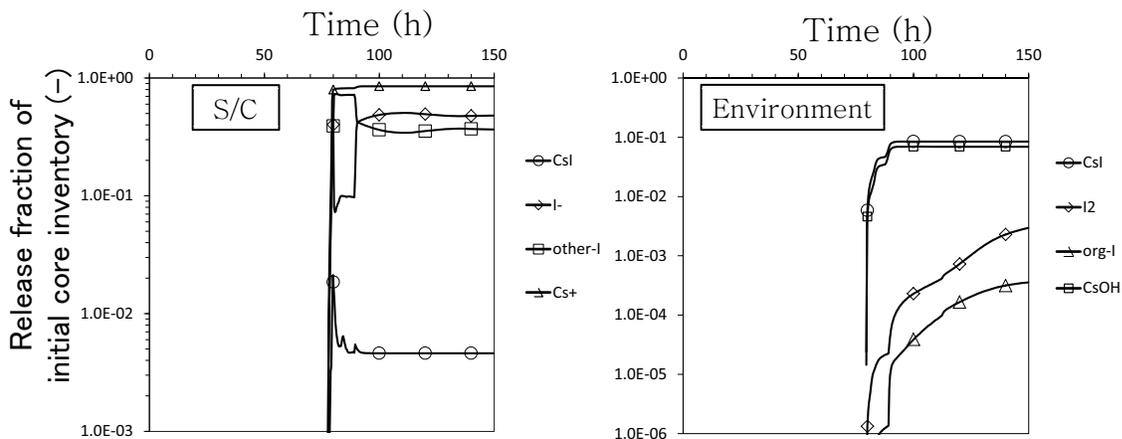
**Figure 12. CsI fraction distribution of unit 2 (DWF case)**

Figures 13 and 14 shows the predicted distributions of the FP species at the S/C and released into the environment in DWF and SCF cases, respectively.

A large difference was not found between DWF and SCF cases for the release fraction of CsI into the environment. This was because the scrubbing effect in the S/C water phase was not significant in the SCF case under a saturated water condition. With regard to the release of volatile iodine ( $I_2$  and org-I), approximately twice larger amount was predicted to be released into the environment in SCF case. However, it should be noted that further studies are necessary in order to significantly reduce uncertainties in modeling for iodine source term such as scrubbing under a high temperature condition, aqueous phase iodine chemistry including influences of seawater and gas-liquid mass transfer characteristics under an expected two-phase flow condition.



**Figure 13. Release fraction of FP in S/C and environment (unit 2, DWF)**



**Figure 14. Release fraction of FP in S/C and environment (unit 2, SCF)**

#### 4. Conclusions

In the present study, the analysis of the accident at Fukushima Dai-ichi NPS has been performed with an integrated severe accident analysis code THALES2 coupled with a detailed iodine chemistry analysis code KICHE for purpose of increasing knowledge on the accident progression and source term. The analysis conditions were mainly set on the basis of information from the OECD/NEA BSAF project.

The depressurization of the RCS in unit 1 was well predicted by taking into account TIP tubes connecting the core region with DW, which failed thermally during the process of core melt progression before the melt-through of the lower head of the reactor vessel. The pressure transients in RCS and DW of unit 2 were reasonably reproduced in case of considering the continuous activation of the RCIC, the intentional opening of the SRVs, the leakage through the DW top flange and the failure of the DW or S/C. The analysis indicated that the whole core materials slumped into the pedestal area of the DW for unit 1. Although a similar result was obtained for unit 2, the core melt progression including melt relocation and lower head failure was anticipated to strongly depend on the water injection rate by fire engine pumps. The total amount of hydrogen generated during the core melt progression was evaluated to be approximately 1,200 kg for both units, corresponding to almost 100 % oxidation of zirconium by steam for unit 1 and approximately 65 % for unit 2.

The prediction indicated that the release fraction of cesium and iodine into the environment was at the order of  $10^{-3}$  for unit 1 and  $10^{-1}$  for unit 2, and the most fraction of both FPs was retained in the water phase of the S/C due to the pool scrubbing effect. The accumulation of iodine in the water phase implied the importance of the iodine chemistry to form volatile iodine such as  $I_2$  and organic iodine and their volatile iodine into the environment were predicted. However, the present analysis showed that the dominant form of iodine to be released into the environment was CsI. It was also found that there was no significant difference in the iodine source term between cases with DW failure and S/C failure for unit 2.

It is supposed that uncertainty analysis is necessary for the iodine source term since it is contributed by various uncertain modeling such as scrubbing, iodine chemistry in an aqueous and gas-liquid mass transfer of volatile iodine.

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