Verification of water radiolysis model in MAAP5.06

Seiji Yamasaki*[a], Yutaka Yoshida [b], Hiromasa Chitose [c]
[a] Hitachi GE Nuclear Energy, Ltd., Hitachi-shi, Japan, seiji.yamasaki.ee@hitachi.com
[b] Hitachi GE Nuclear Energy, Ltd., Hitachi-shi, Japan, yutaka.yoshida.ka@hitachi.com
[c] Hitachi GE Nuclear Energy, Ltd., Hitachi-shi, Japan, hiromasa.chitose.cy@hitachi.com

Abstract: The licensing review under the new regulatory standards in Japan requires measures to prevent containment failure due to hydrogen combustion in the containment vessel in the event of a severe accident. In BWRs, the containment atmosphere is replaced with nitrogen to prevent hydrogen and oxygen concentrations in the containment from reaching flammable limits, even if hydrogen gas is generated due to zirconium-water reactions. It is also important to consider the gradual increase of oxygen concentration in the containment vessel due to water radiolysis in the long term. Since the conventional MAAP code (ver.4) did not include a water radiolysis model, the hydrogen and oxygen concentrations in the containment vessel were calculated by post-processing the results of the MAAP analysis to account for water radiolysis, and it was confirmed that these did not reach the flammable limit. On the other hand, the latest MAAP code (ver.5.06) has a new G-value-based water radiolysis model applied in the Japanese licensing, which allows the calculation of hydrogen and oxygen concentrations that account for water radiolysis directly within the code. Therefore, the model was verified in this study by comparing it with the conventional post-processing method. Two scenarios were compared for model validation to facilitate analysis of the causes of the differences. The first scenario is the In-vessel scenario of TQUV (transient with loss of all ECCS injections), which allows comparison of hydrogen and oxygen production from water radiolysis in the reactor. The second scenario is the Ex-vessel scenario of TQUV, which allows comparison of hydrogen and oxygen production from water radiolysis not only in the reactor, but also in FP released from the damaged core and in molten core released into the containment vessel. As a result of the comparison, it was confirmed that the newly added water radiolysis model in MAAP5.06 is valid, since the two models are in close agreement.

Keywords: severe accident, BWR, G-value-based water radiolysis, MAAP5, model verification.

1. INTRODUCTION

Since the Fukushima Daiichi nuclear accident, new regulatory requirements [1] have been established in Japan, requiring the effectiveness evaluations for severe accident countermeasures. These evaluations require applicants for the Permission of Reactor Installment License to conduct analyses and confirm the effectiveness of these countermeasures (Article 37 of the Regulations on the Permission of Reactor Installment License). Specifically, in the event of an accident that could lead to a severe accident, the applicant must confirm that the necessary measures to prevent severe damage to the reactor core are effective (interpretation of Paragraph 1 of the same Article). Additionally, even in the event of a severe accident, they must confirm the effectiveness of measures to prevent damage to the primary containment vessel and the release of radioactive materials at abnormal levels outside the plant (interpretation of Paragraph 2 of the same Article).

The interpretation of Paragraph 2 of Article 37 requires that the effectiveness of measures to prevent damage to the containment vessel be confirmed for each failure mode of the containment vessel. A containment vessel failure mode is a classification of events that could lead to damage to the containment vessel and the release of radioactive materials at abnormal levels outside the facility after significant core damage, focusing on the type of load to the containment vessel. Comprehensive identification of these failure modes is necessary as a prerequisite for effectiveness evaluation. Based on previous research, a "containment vessel failure mode that is always assumed" has been defined. Specifically, this includes static load due to atmospheric pressure and temperature (overpressure and overtemperature damage), high-pressure melt release/direct heating of the containment vessel atmosphere, molten fuel-coolant interaction outside the reactor pressure vessel, hydrogen combustion, direct containment vessel contact (shell attack), and molten core-concrete interaction.

In the "hydrogen combustion", the hydrogen concentration in the primary containment vessel (PCV) increases due to hydrogen gas generated by the zirconium-water reaction, radiolysis of water, metal corrosion, molten core-concrete interaction, etc. The oxygen concentration in the primary containment vessel also increases due to the radiolysis of water. If no mitigation measures are taken, the hydrogen gas and the oxygen gas in the PCV can react, causing severe combustion and potentially leading to damage to the PCV. Therefore, in Boiling Water Reactors (BWRs), in addition to inerting the atmosphere inside the PCV by replacing it with nitrogen gas, nitrogen injection into the PCV via a portable nitrogen supply unit prevents the hydrogen and oxygen concentrations from reaching flammable limit, thereby preventing damage to the PCV.

In these effectiveness evaluations, the MAAP (Modular Accident Analysis Program) code is used as an analytical tool to evaluate the behavior of the containment vessel during a severe accident. The MAAP code, owned by EPRI, analyzes the release and transfer behavior of thermal-hydraulic and fission products in the reactor pressure vessel and containment vessel during a severe accident in a light water reactor [2]. MAAP includes models for significant accident phenomena that may occur in the reactor core, reactor pressure vessel, and primary containment vessel, allowing for the evaluation of hydrogen gas generation from zirconium-water reactions and molten core-concrete interactions.

However, MAAP Ver. 4, used in the effectiveness evaluations, did not include a model for the radiolysis of water within the code, making it impossible to evaluate the gas generation from water radiolysis in the containment vessel directly. Consequently, external post-processing of MAAP analysis results was used to assess gas phase concentrations in the containment vessel, ensuring they remained below flammable limit.

On the other hand, the latest MAAP ver. 5.06 incorporates a radiolysis model for water, enabling the calculation of gas generation from water radiolysis within the MAAP code without post-processing. Verification of this model is important for applying it to effectiveness evaluations for the failure mode of “hydrogen combustion.”

This paper compares the results from traditional post-processing methods with the radiolysis model in MAAP5.06 to validate its accuracy and examine the model's suitability for regulatory applications.

2. EFFECTIVENESS EVALUATION FOR THE FAILURE MODE OF HYDROGEN COMBUSTION

2.1. Selection of Accident Sequences

In effectiveness evaluations, accident sequences are selected for each containment vessel failure mode. Specifically, for each failure mode, the plant damage state (PDS) considered most severe and most likely to lead to the failure mode is identified. From the accident sequences, the most severe sequence from the viewpoint of the failure mode is selected.

In BWRs, nitrogen is used to replace the atmosphere in the primary containment vessel, maintaining a low initial oxygen concentration. With core damage, the hydrogen concentration can easily exceed 13 vol%. Therefore, keeping the oxygen concentration low is critical for preventing hydrogen combustion.
Oxygen gas is generated by the radiolysis of water. However, the oxygen concentration is influenced by the presence of other gases. We focus on the hydrogen gas generation by the zirconium-water reaction which is expected to have a significant impact on the gas composition in the primary containment vessel after core damaged.

The behavior of the zirconium-water reaction when reactor water injection is not expected can be divided into large LOCA and other PDS based on the release route of the coolant outside the reactor pressure vessel at the time of the event.

In large LOCA, the reactor pressure vessel is immediately depressurized at the event, and a large amount of coolant is discharged outside the vessel. As a result, the amount of coolant contributing to the zirconium-water reaction is reduced. Although the hydrogen concentration exceeds 13 vol%, the amount of hydrogen gas generated is considered to be lower than in other PDS. Therefore, it is possible that the oxygen concentration increased by the radiolysis of water in large LOCA is higher than in other PDS.

Furthermore, considering the influence of the presence or absence of damage to the reactor pressure vessel, it is appropriate to assume that the same PDS does not lead to damage to the reactor pressure vessel because the non-condensable gas generated by the molten core-concrete interaction in the pedestal may contribute to lowering the oxygen concentration when the reactor pressure vessel is damaged.

From the above, an accident sequence is identified in which a severe accident occurs, involving both large LOCA and the loss of emergency core cooling system injection function, and the reactor pressure vessel damage is avoided by the alternative injection system.

2.2. Implementation of Severe Accident Analysis

Severe accident analysis is conducted on the accident sequence extracted in Section 2.1. The MAAP code is utilized for the analysis.

MAAP is a code used to analyze the thermohydraulic and radioactive material behavior in a plant from core damage to the release of radioactive materials into the environment for accident sequences involving core damage.

After core damage, the inside of the reactor and the primary containment vessel are divided into primary systems, drywell, and wetwell. Various phenomena such as core heat-up, oxidation and rupture of fuel cladding, molten core transfer behavior and cooling, generation of hydrogen gas and water vapor, molten core-concrete interaction, containment vessel pressure and temperature, release of radioactive materials, and transfer/deposition behavior are modeled to evaluate plant behavior during severe accidents.

2.3. Consideration of radiolysis of water by post-processing

2.3.1. Radiolysis model of water

When water absorbs radiation energy such as γ rays, radiolysis of water occurs in a very short period, producing H (hydrogen atoms), OH radicals, e^{aq-} (hydration electrons), HO_2 radicals, H^+ (hydrogen ions) and molecular products H_2 and H_2O_2 (hydrogen peroxide). In addition, oxygen is produced by the decomposition of hydrogen peroxide.

Because MAAP4 code does not have the model of radiolysis of water, it is necessary to separately evaluate the generation of hydrogen gas and oxygen gas by radiolysis of water in the effectiveness evaluation in the failure mode "hydrogen combustion".

In BWRs, the primary containment vessel is replaced by nitrogen gas in during the operation.
When the reactor core is damage and after the reactor pressure vessel is damaged, the dominant production processes of hydrogen gas are such as the zirconium-water reaction and the molten core concrete interaction. For oxygen gas, the dominant production process is the radiolysis of water.

From the viewpoint to keep the concentration of gases in PCV below flammable limit, it is important to keep the concentration of oxygen gas low rather than the concentration of hydrogen gas which is exceeding the limit in relatively short period by the above reactions.
Equation (2-1) shows the evaluation formula for gas generated by the radiolysis model of water based on the G value.

$$\Delta n = A \times Q_{decay} \times E \times G \times \Delta t \qquad (2-1)$$

Here, Δn is the amount of hydrogen or oxygen gas generated by the radiolysis of water [mol], A is a proportionality constant for unit conversion, Q_{decay} is decay heat [W], E is radiation absorption rate [-], G is the effective G value [molecule / 100 eV], and Δt is the elapsed time [sec].
The radiation absorption ratio in the reactor vessel was set to 10% conservatively from the analysis result that the absorption rate of radiation emitted from the core into the water was about 1%.
In addition, considering that the FP outside the reactor vessel is dispersed in water, 100% of the radiation energy contributes to the radiolysis of water.

The number of atoms and molecules produced per 100 eV of absorbed energy of radiation is called the G value. The G value has “initial G value,” considering only the effect of the radiolysis of water, and “effective G value,” considering not only the effect of the radiolysis of water but also the effect of the chemical reaction in which the products of radiolysis recombine and return to water molecules.
After irradiation begins, the product of radiolysis increases and recombine reaction of the products to water increases according to the concentration of the product. As a result, the production ratio of hydrogen and oxygen molecules gradually decreases.

The trend of the relationship between hydrogen and oxygen concentration and the absorbed dose of water assumes a curve in which the increase in the concentration is gradually suppressed, rather than a temporary peak of increase in the concentration.

From the viewpoint of evaluating the macroscopic phenomenon of increased concentrations in the PCV, it is appropriate to use the effective G value including the effect of chemical reactions such as recombination.
In previous studies on G values [3], the evaluation results of G values for various pH, water temperature, water quality and irradiation dose were reported by simulating the environment inside the containment vessel of the actual containment vessel. The G values of hydrogen and oxygen are set to $G(H_2) = 0.06$ and $G(O_2) = 0.03$ as values based on these research results.

2.3.2. Method for Evaluating Hydrogen and Oxygen Concentrations in PCV

The method for evaluating hydrogen and oxygen concentrations in the primary containment vessel considering the effect of radiolysis is as follows. Figure 2-1 shows the flow of evaluation of hydrogen and oxygen concentrations in the primary containment vessel.

- Calculate the initial number of moles of oxygen gas and nitrogen gas in the drywell and wetwell from the number of moles of nitrogen gas when the initial oxygen concentration of the primary containment vessel is FO2 [vol%], using the nitrogen gas moles in the drywell and wetwell obtained from MAAP analysis.
- From the decay heat in the reactor vessel, the drywell and wetwell, calculate the amount of oxygen gas and hydrogen gas generated by the radiolysis of water according to Equation 2-1.
- Distribute the hydrogen gas and oxygen gas generated by the radiolysis of water between the drywell and the wetwell according to the amount of transfer evaluated based on MAAP results.

The radiolysis model of water based on the G value implemented in MAAP5.06 can directly analyze the number of moles of gas in the containment vessel according to the model based on Equation 2-1. This allows the effect of gases generated by the radiolysis of water to be considered in severe accident analysis.

2.3. Comparison with Criteria in Effectiveness Evaluation

For the conditions under which hydrogen combustion or detonation occurs, a ternary diagram of hydrogen, air, and water vapor as shown in figure 2-1 is known[4]. Ignition and deflagration can occur at oxygen concentrations of 5% or more, hydrogen concentrations of 4% or more, and steam concentrations of 60% or less, and the flame propagation speed is slow, placing quasi-static loads on the containment vessel. On the other hand, detonation can occur at hydrogen concentrations of 13% or more, and flame acceleration causes the combustion wave to travel at supersonic speed, placing a dynamic load on the containment vessel. The basis for judging the efficacy evaluation for hydrogen combustion is defined as "the hydrogen concentration in the primary containment vessel is 13 Vol% or less or 5% Vol or less in terms of dry conditions"[2].

By comparing the hydrogen and oxygen concentrations in the primary containment vessel obtained in 2.3 with the above basis for judgement, the appropriateness of the effectiveness evaluation for hydrogen combustion is judged.

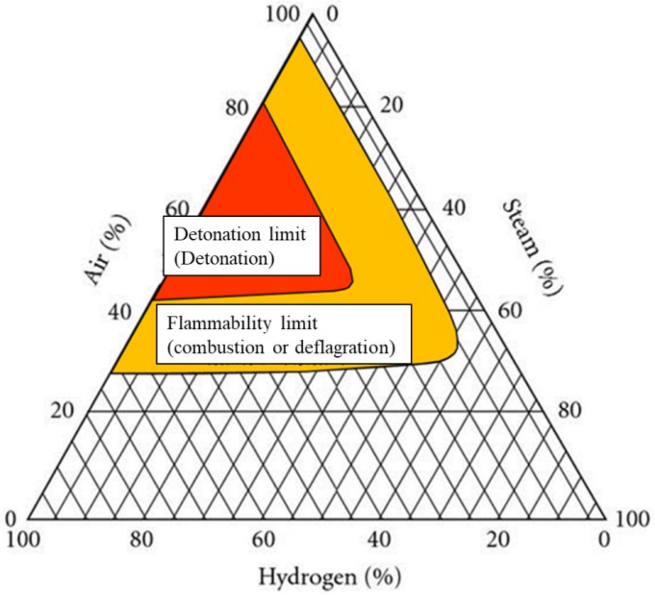


Figure 2-1 Ternary diagram of hydrogen combustion [4]

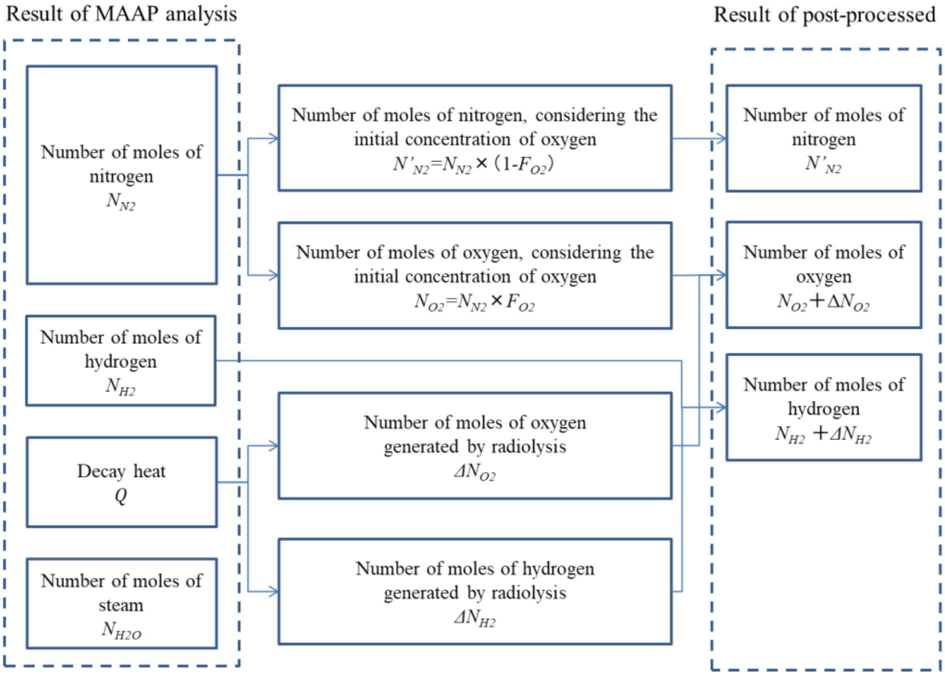


Figure 2-2 Gas Evaluation Flow in Post-Processing method

3. ANALYSIS CONDITIONS FOR VERIFICATION OF RADIOLYSIS MODEL

The analysis conditions are shown in Table 3-1. The analysis target is assumed to be a representative Advanced Boiling Water Reactor (ABWR). As a sequence, "loss of high pressure and low-pressure water injection function (TQUV)" is assumed. Specifically, after the occurrence of a transient event (here, total loss of feedwater flow rate), the high-pressure water injection function (RCIC and HPCF) and the low-pressure water injection function (RHR with low pressure water injection mode) is lost, and the reactor water level decreases due to the release of steam from the safety relief valves. The analysis case compares the amount of hydrogen and oxygen generated and the hydrogen generation rate in the event progression of the following two scenarios.

a. "In-Vessel" scenario

Figure 3-2 shows the accident progress diagram of this scenario.
In this scenario, the radiolysis of water occurs in the core and the FP released from the core since the accident is resolved in the reactor pressure vessel. For this reason, a simple scenario was selected for the verification of the radiolysis model of water.

The main events in this scenario are shown below.

- As an initiating event, a total loss of feed water flow occurs.
- The low reactor water level (level 3) signal is generated and the reactor scrums, but the RCIC fails to start at the reactor water level (level 2), the HPCF fails to start at the low reactor water level (level 1.5), and the RHR (Low Pressure water injection mode) fails to start at the low reactor water level (level 1).
- Approximately 14 minutes after the occurrence of the event, the reactor is rapidly depressurized by remotely opening the eight safety relief valves from the main control room, and the alternative reactor water injection system is started after the reactor depressurized.
- When the reactor rapid depressurization is started, the reactor water level drops due to the outflow of coolant the reactor, and drops to the top of active fuel, and core damage occurs. Thereafter, when water injection by the alternative low pressure water injection system is started, the reactor water level recovers and the reactor core is re-flooded.

b. "Ex-Vessel" scenario

Figure 3-3 shows the accident progress diagram of this scenario.
In this scenario, the radiolysis of water occurs not only in the core but also in the melting core under the reactor pressure vessel, in the melting core the containment vessel, and in the FP released from the core because the reactor pressure vessel breaks and the molten core falls into the lower reactor pressure vessel. For this reason, in the verification of the radiolysis model of water, it was selected as a scenario for confirming the validity of the model at all positions where the radiolysis of water could occur.

The main events in this scenario are shown below.

- As an initiating event, a total loss of feed water flow occurs.
- The low reactor water level (level 3) signal is generated and the reactor scrums, but the RCIC fails to start at the reactor water level (level 2), the HPCF fails to start at the low reactor water level (level 1.5), and the RHR (Low Pressure water injection mode) fails to start at the low reactor water level 1).
- When the reactor water level reaches a position 10% above the length of the active fuel length from the bottom of the fuel, the reactor is rapidly depressurized by remotely opening the two safety relief valves from the main control room.
- When the temperature of the bottom of the containment vessel reaches at 300°C, water-filling into the lower PCV by alternative water injection system starts before the reactor pressure vessel is damaged.
- Since it is assumed that the reactor water injection by an alternative low pressure water injection system cannot be carried out after the reactor depressurization, the reactor pressure vessel will be damaged.
- When the reactor pressure vessel is damaged and the molten core fall into water with a water level of about 2m depth pool under the containment vessel, heat is transferred from the melt core to the water pool in the lower PCV, and the pressure rises due to the generation of water vapor.

Table 3-1 Analysis conditions

	Value	Remarks
Analysis code	MAAP5.06	-
Thermal Output	3,926MWt	Rated reactor heat output
Reactor Pressure	7.07MPa[gage]	Rated reactor pressure
Reactor water level	Normal operating water level (+119 cm from the bottom edge of the separator skirt)	Water level in normal operation
Reactor water flow	52,500t/h	Rated water flow
Fuel type	9×9 Fuel (Type A)	-
Decay heat	ANSI/ANS5.1-1979 33GWd/t burnup	Considering the variability of the degree of combustion at the end of the cycle, set for 10% maintainability
Volume of containment vessel (Dry well)	7,350m^3	Design value of the volume in the dry well (total volume minus the volume of internal equipment and structures)
Volume of containment vessel (wet well)	Vapor phase: 5,960m^3 Liquid phase: 3,580m^3	Design value of the volume in the wet well (excluding the volume of internal equipment and structures)
Vacuum breaker	3.43 kPa (Drywell-Wetwell Differential Pressure)	Vacuum breakdown device setpoint
Wetwell pool water level	7.05m (normal operating water level)	Set as the wetwell pool water level during normal operation
Wetwell pool temperature	35℃	Set as the upper limit of the wetwell pool water temperature during normal operation
Containment vessel pressure	5.2kPa[gage]	Set as containment vessel pressure during normal operation
Containment vessel temperature	57℃	Set as the containment vessel temperature during normal operation
G value on severe accident	H2: 0.06 O2: 0.03	Values based on previous studies

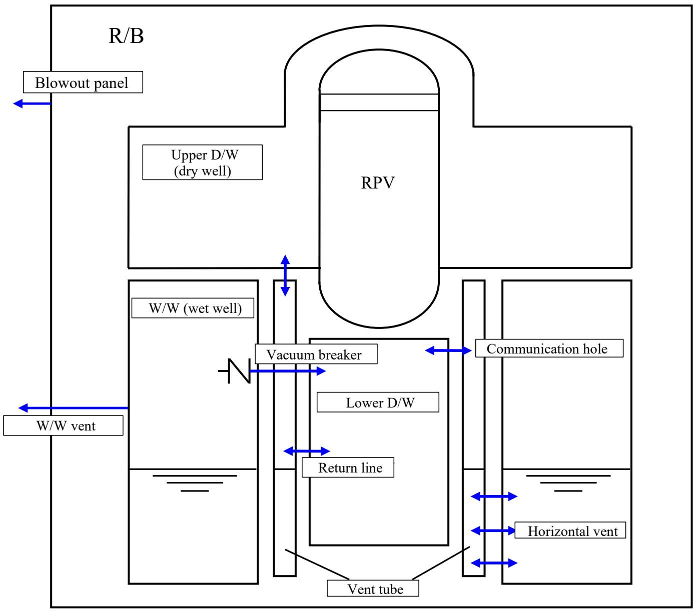
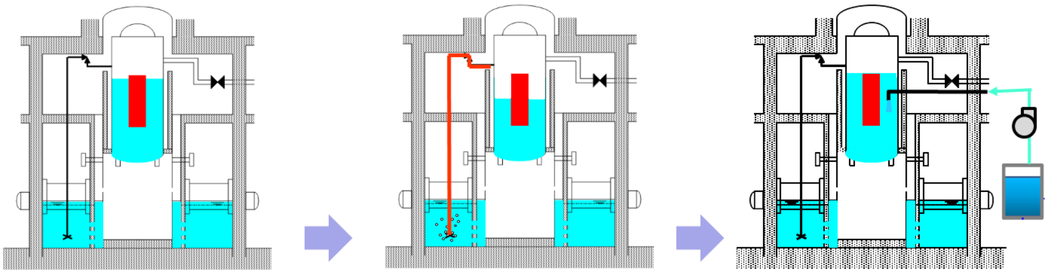


Figure 3-1 Nodalization of the ABWR Plant in MAAP Code

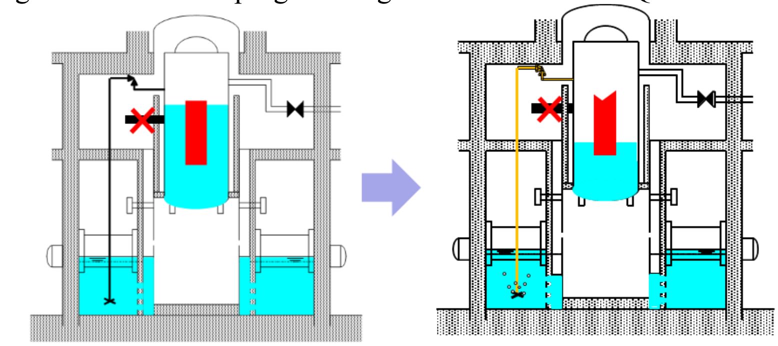


Accident occurred (Loss of feedwater)

Core exposed by decompression with SRV opened

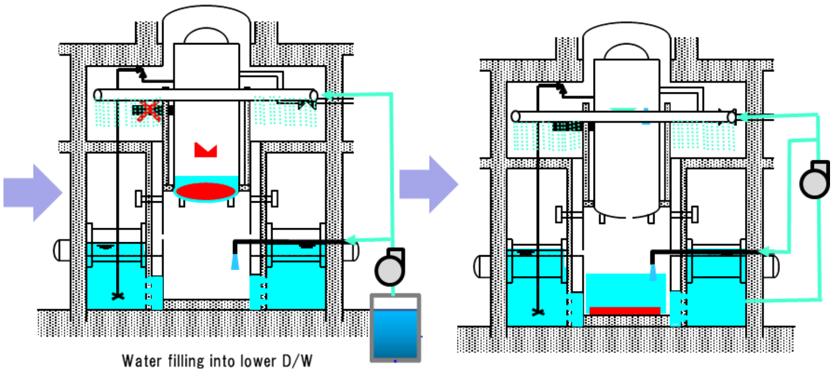
Core reflooded with low pressure alternative injection system

Figure 3-2 Accident progress diagram in "In-vessel" TQUV Scenario



Accident occurred (Loss of feedwater)

Core damaged by lowering water level of decompression with SRV opened



Water filling into lower D/W

Core dropped from RPV to lower D/W

Figure 3-3 Accident progress diagram in "Ex-vessel" TQUV Scenario

4. ANALYSIS RESULTS FOR VERIFICATION OF RADIOLYSIS MODEL

a. "In-vessel" scenario

Figure 4-1 shows a graph of the accumulated amount of oxygen and hydrogen generated in this scenario, and figure 4-2 shows the amount of hydrogen generated in each region.

As shown in Figure 4-1, the accumulated amount of hydrogen and oxygen generated by the radiolysis of water was almost identical between the MAAP4 with post-processing and the MAAP5 radiolysis model. Given that the ratio of hydrogen and oxygen is 2:1, the results of this scenario are considered reasonable.

As shown in Figure 4-2, the hydrogen generation rate generated by RPV, and W/W was also consistent with the MAAP4 with post-processing and the MAAP5 radiolysis model. In RPV, it can be confirmed that the hydrogen generation rate due to the radiolysis of water decreases rapidly as the decay heat decays.

A slight hydrogen generation is observed in W/W about 0.8 hours after the event. This is due to the transition of the FP from RPV to W/W via safety relief valves after the core damage.

b. "Ex-vessel" scenario

Figure 4-3 shows a graph of the accumulated amount of oxygen and hydrogen generated moles in this scenario, and figure 4-4 shows the hydrogen generation amount in each region.

As shown in Figure 4-3, the results of the MAAP4 with post-processing and the MAAP5 radiolysis model were in good agreement in this scenario as well. Given that the ratio of hydrogen and oxygen is 2:1, the results of this scenario are considered reasonable. The ratio of hydrogen and oxygen generation is 2: 1, and the results are considered reasonable.

As shown in Figure 4-4, the hydrogen generation rates generated by RPV, D/W, and W/W were almost identical between the MAAP4 with post-processing and the MAAP5 radiolysis model. The rate of hydrogen generation in D/W is sometimes higher in the MAAP4 with post-processing than in the MAAP5 radiolysis model, this is due to the conservative assumption of the method of MAAP4 with post-processing that radiolysis of water is occurred even in the case of no water in RPV or D/W.

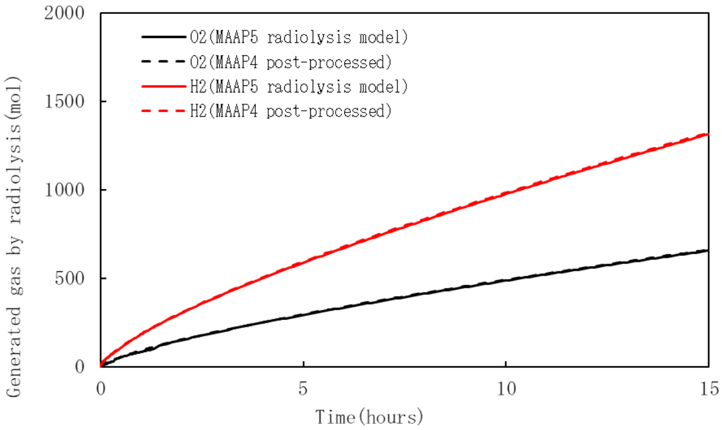


Figure 4-1 Amount of oxygen and hydrogen generated in the "In-vessel" scenario.

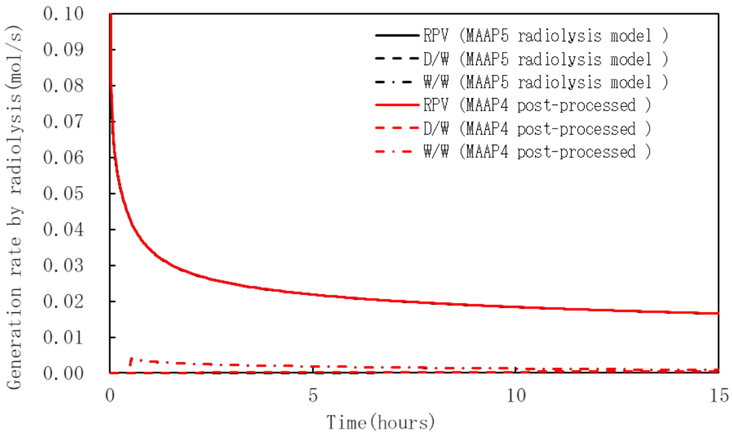


Figure 4-**2** Comparison of Hydrogen Evolution Rates in the "In-vessel" Scenario

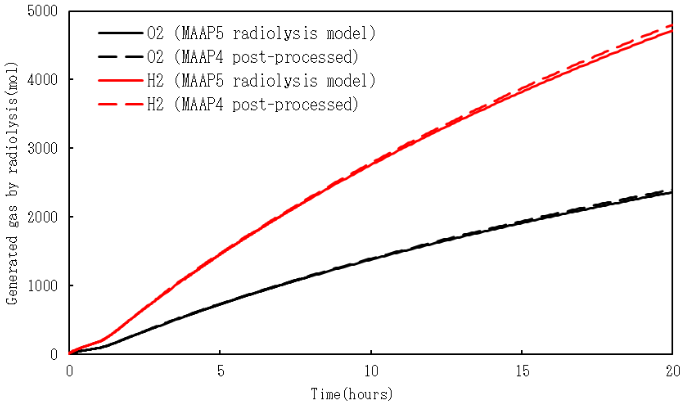


Figure 4-3 Amount of oxygen and hydrogen generated in the "Ex-vessel" scenario.

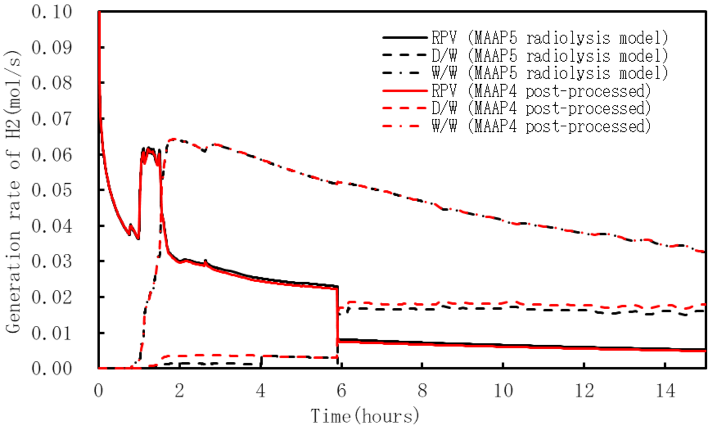


Figure 4-4 Comparison of Hydrogen Evolution Rates in the "Ex-vessel" Scenario

5. CONCLUSION

In this paper, in order to confirm the validity of the newly implemented radiolysis model in MAAP5, we compared the results of the analysis results of MAAP4, which has been used to evaluate the effectiveness of countermeasures for the failure mode of “hydrogen combustion”, with the results of the radiolysis model incorporated in MAAP5. As a result of the comparison, the two sides showed a good agreement, and the validity of the radiolysis model of water in MAAP 5 is judged to be reasonable.

By using the radiolysis model of water embedded in MAAP5, we now are available to directly and simultaneously evaluate the changes of the concentration of gases and the pressure in the PCV due to the radiolysis of water, which the conventional MAAP4 with post-processing method was not able to evaluate.

We consider it useful to apply the radiolysis model of MAAP5 to the effectiveness evaluation of the failure mode of “hydrogen combustion”.

References

[1] Nuclear Regulation Authority, “New Regulatory Requirements for Light-Water Nuclear Power Plants” (in Japanese), (2013).
[2] Toshiba Energy Systems & Solutions Corporation and Hitachi-GE Nuclear Energy, Ltd., “Boiling Water Reactor Nuclear Power Plants --- Modular Accident Analysis Program (MAAP) for Evaluating the Effectiveness of Measures Against Severe Accidents” (in Japanese), TLR-094, HLR-123, (2018).
[3] H. Ito, et al., “Ratio of Combustible Gas Generation by Radiolysis of Water at the Time of Accident (I)” (in Japanese), Atomic Energy Society of Japan, B18 “Annual meeting of Autumn 1987”, (1987).
[4] B. R. Sehgal, “Nuclear Safety in Light Water Reactors,” (2012).