Performance Simulation of a Catalyst in a Passive Radioactive Material Reduction System

Jongtae Kim [a*], Keun Sang Choi [a], Jaehoon Jung [a], Youngsu Na [a]
[a] *KAERI, Daeduk-daero 989-111, Daejeon, Korea*
[*] *Corresponding author: ex-kjt@kaeri.re.kr*

Keywords : Fission Product Filtering System, Catalyst, Containment, Severe Accident, Contain-3D

1. Introduction

The most important factor in preventing the release of large amounts of radioactive material into the external environment of the containment building during a severe accident is to ensure the integrity of the containment building. Another method is to actively reduce the concentration of radioactive fission products suspended in the containment atmosphere. The most active method of removing fission products inside the containment is the operation of the spray system installed in a nuclear reactor containment. However, this method can rapidly lower the composition of the containment atmosphere, especially the concentration of steam, thereby relatively increasing the concentration of hydrogen. Moreover, since it is an active system that requires power, additional power sources are necessary in the event of a station blackout accident. To compensate for the shortcomings of radioactive fission products reduction by spraying, a fully passive fission product reduction device is being developed. This device uses a hydrogen catalyst as a method to draw radioactive substances into a filter. In this paper, experimental and analytical research results for evaluating the performance of the hydrogen catalyst are presented.

2. Methods and Results

Catalytic filtering system shown in Fig. 1 consists of a square-duct housing, an aerosol filter, an iodine absorbent, and a catalyst. The catalyst induces a buoyant flow for aerosol filtration through the catalytic reaction of hydrogen and oxygen.

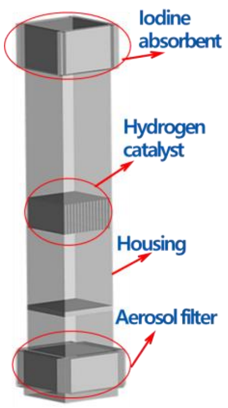

Fig. 1. Schematic of Passive Radioactive Material Reduction System.

To evaluate hydrogen recombination and flow induction, a simplified test device comprising only a housing and catalytic body was constructed and installed in the SPARC test facility [1].

2.1 Experimental Method and Conditions

The SPARC test facility consists of a pressure vessel, steam and hydrogen injectors, and sensors for measuring gas species concentrations and temperature. The configuration of the catalytic filtering system and the hydrogen injection nozzle ring are shown in Fig. 2. Gas species such as hydrogen, oxygen, and water vapor were measured at 14 probe locations.

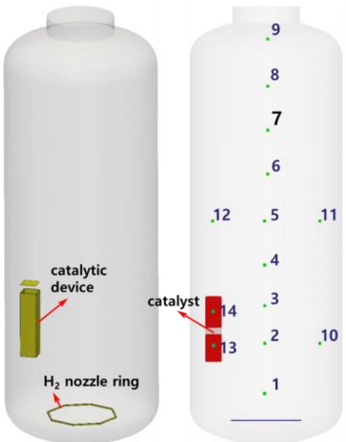

Fig. 2. Catalyst test facility and measurement points of gas concentrations and temperature.

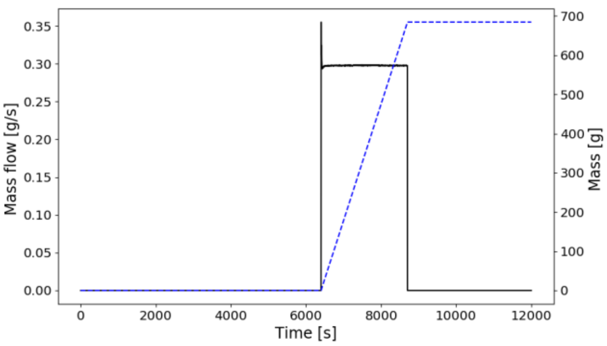

Fig. 3. Injection rate and total mass of hydrogen for the catalyst test.

At the beginning of hydrogen injection, the vessel conditions were:

- Pressure: 1.329 bar
- Temperature: 64 ℃
- Water vapor concentration: 12% (100% relative humidity).

The hydrogen injection rate and total injected mass are shown in Fig. 3.

2.2 Catalytic Reaction Modeling

In general, the hydrogen recombination rate of a PAR (Passive Autocatalytic Recombiner) is expressed through empirical correlation equations, as in Eq. (1):

$$R = correlation(p,\ T,\ x_{h2},\ x_{o2},\ x_{h2o}) \tag{1}$$

Because the catalytic system in this study is still under development, no established correlation exists for its performance. Instead, a diffusion-based PAR model [2,3] was applied, which can in principle be used for any catalyst type. Verification against experimental data is, however, essential.

The mass diffusion coefficient of each gas species at the catalytic surface was obtained using Sherwood number correlations. The hydrogen oxidation process involves multiple reaction steps, but in this study, diffusion was assumed to dominate because it is slower than the chemical reaction rates.

Accordingly, the hydrogen and oxygen removal rates were expressed as diffusion-limited mass fluxes (Eqs. 2–3).

$$\phi_{H_2} = \rho h_{m,H_2} Y_{H_2} \tag{2}$$

$$\phi_{O_2} = \rho h_{m,O_2} Y_{O_2} \tag{3}$$

The diffusion coefficient h_m of the gas component is obtained using the Sherwood number correlation as follows. The Sherwood number correlation is expressed as a function of the Gr number in the case of natural convection and as a function of the Re number in the case of forced convection, depending on the flow characteristics. Here, since three-dimensional modeling is performed for the catalyst reaction region and the housing region, the Sherwood number correlation based on the Reynolds (Re) and Schmidt (Sc) numbers, as in Eq. (6), is used.

$$Re_L = \rho_g U_g L_{par}/\mu_g \tag{4}$$

$$Sc = \nu_g/D \tag{5}$$

$$Sh = \frac{h_m L_{par}}{D} = \begin{cases} 0.664 Re_L^{1/2} Sc_{H_2}^{1/3}, & Re_L < 5\times10^5 \\ 0.037 Re_L^{4/5} Sc_{H_2}^{1/3}, & Re_L > 5\times10^5 \end{cases} \tag{6}$$

2.3 Experimental Results

In the test, hydrogen concentration was continuously increased in the test vessel during the injection of hydrogen. Fig. 4 shows the measured hydrogen concentrations over time at the 14 probing locations. The hydrogen mass removed by the catalyst can be calculated by two methods. Method-1 is based on the inlet and outlet hydrogen mass flow rates of the PAR housing and Method-2 is by evaluating the decreasing rate of the remaining hydrogen inside the pressure vessel.

Due to possible non-uniformity of gas concentrations and limited probe locations, both methods are subject to measurement errors. For example, Method 1 may overestimate removal because only one probe was placed at the housing inlet and outlet. Method 2 is more reliable in later stages, when hydrogen is well mixed.

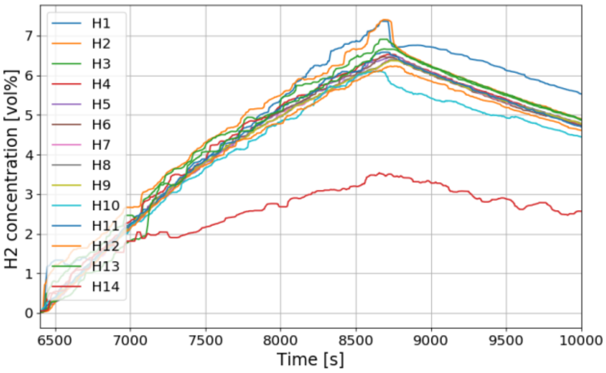

Fig. 4. Measured hydrogen concentrations over time.

After finishing the hydrogen injection, the injected hydrogen is continuously diffused and removed by the catalytic reaction. So, the distribution of hydrogen in the test vessel becomes very uniform at the final stage of the experiment. The amount of the remaining hydrogen at the end of the test is used as a reference point for tuning the amount of the removed hydrogen evaluated by Method-1.

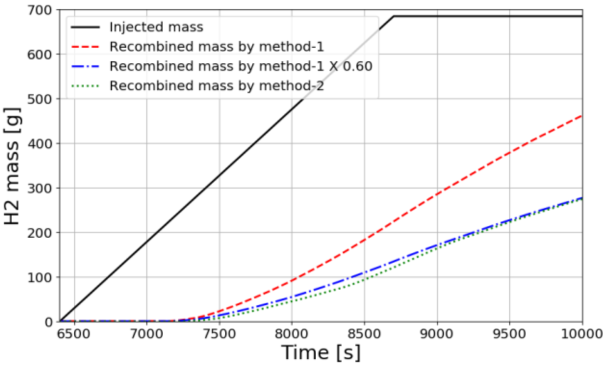

Fig. 5. Removed hydrogen masses over time evaluated by Method-1 and Method-2.

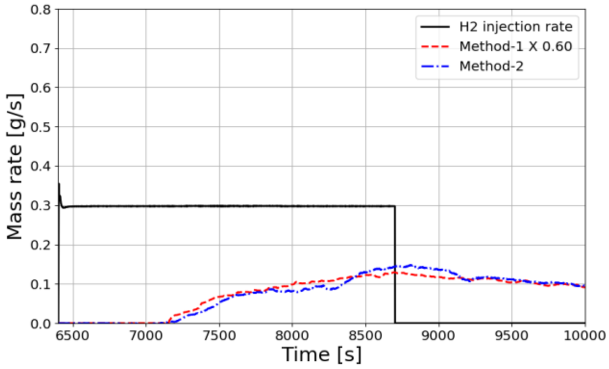

Fig. 6. Hydrogen removal rates by Method-1 and Method-2

Comparisons of hydrogen removal using both methods are shown in Figs. 5 and 6. After applying a correction factor (0.6) to Method 1, the two methods agreed closely.

2.4 Analysis Results

Fig. 7 shows the hydrogen distributions simulated by the Contain-3D code [4]. The buoyant jet of hydrogen injected from the nozzle ring and the recombined hot gas plume are shown in the figure.

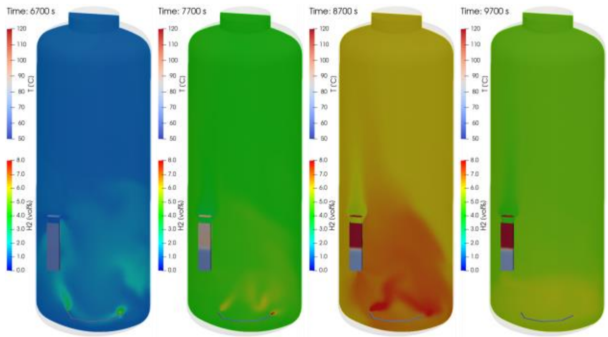

Fig. 7. Simulated hydrogen distributions over time.

The hydrogen concentrations along the center line of the test vessel were compared between the simulated and test results in Fig. 8. Increasing and decreasing characteristics of the hydrogen concentrations in the test were well predicted.

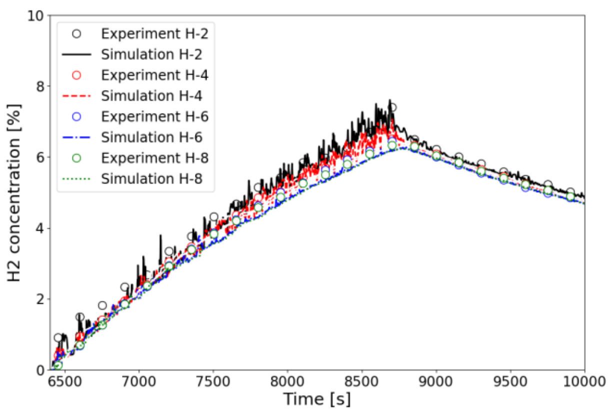

Fig. 8. Comparisons of hydrogen concentrations over time at the center line of the test vessel.

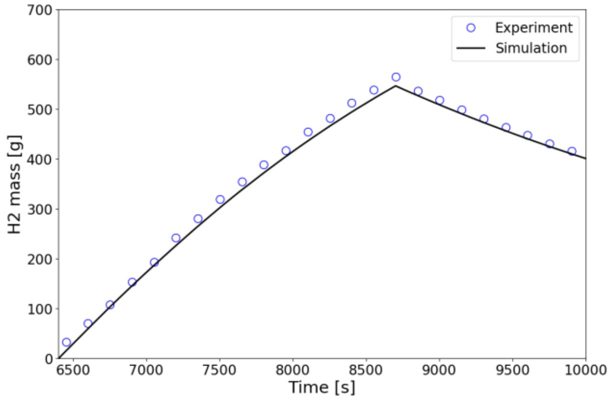

Fig. 9. Comparison of remaining hydrogen mass over time.

The hydrogen mass inventory in the test vessel increases during hydrogen injection and decreases by the catalyst after finishing the injection. In Fig. 9, the integrated mass of hydrogen remaining in the test vessel is plotted over time. The numerical result is in good agreement with the test result.

In the experiment, pressure in the test vessel is slowly increasing during the hydrogen injection (Fig. 10). In general, PAR operation can increase gas pressure by its exothermic reaction. In the current test, the vessel pressure remained nearly constant after hydrogen injection. This was attributed to wall condensation of steam, as confirmed by the simulation.

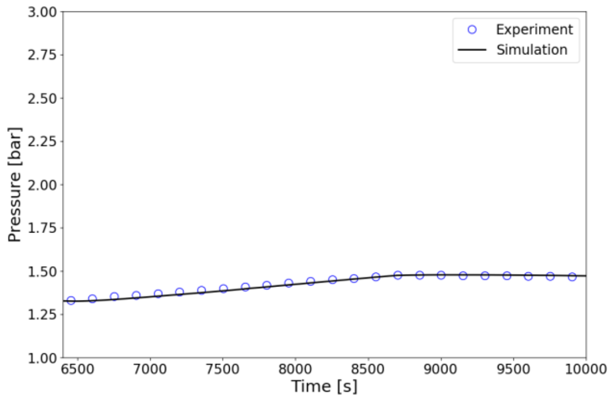

Fig. 10. Comparison of vessel pressure over time.

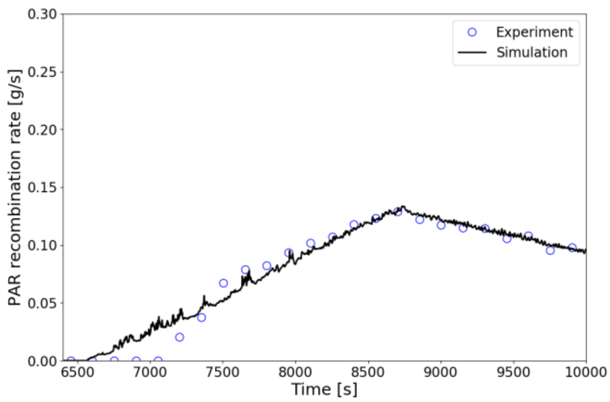

Fig. 11. Comparison of hydrogen removal rate over time.

Finally, Fig. 11 compares the hydrogen removal rate obtained experimentally and numerically. The diffusion-based catalytic model reproduced the observed removal rates accurately.

3. Conclusions

Currently, a fully passive fission product reduction device is being developed, which generates flow in the housing by a catalytic body. Its hydrogen removal performance was tested at the SPARC facility and validated against numerical simulations. The diffusion-based catalytic reaction model implemented in Contain-3D showed good agreement with experimental results, demonstrating predictive capability.

Future work should focus on improving catalyst efficiency to enhance hydrogen removal rates.

ACKNOWLEDGMENTS

This research was supported by the Korea Institute of Energy Technology Evaluation and Planning (KETEP) grant funded by the Korea government (MOTIE, Ministry of Trade, Industry & Energy). (RS-2025-02318177)

REFERENCES

[1] J. Kim and J. Jung, Evaluation of Hydrogen Recombination Characteristics of a PAR using SPARC PAR Experimental Results, Vol. 55, p.4382, 2023

[2] B. Gera, V. Verma, and J. Chattopadhyay, Development and validation of diffusion based CFD model for modelling of hydrogen and carbon monoxide recombination in passive autocatalytic recombiner, Vol. 55, p.3194, 2023.

[3] J. Kim, J. Jung, and S.H. Kim, CFD Modeling for Hydrogen Removal by a Passive Autocatalytic Recombiner in Oxygen-deficient Conditions, Vol. 51, No. 1, 2025

[4] J. Kim and K. Lim, Validation and Application of Code for Three-Dimensional Analysis of Hydrogen–Steam Behavior in a Nuclear Reactor Containment during Severe Accidents, applied science Vol.14, No.15, 2024

.