An Experimental Study on Exothermic Characteristics of Catalyst for Passive Airborne Radioactive Material Reduction System

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1. Introduction

In the event of a severe accident, a substantial quantity of reactor coolant and radioactive material may be released into the atmosphere of the containment. Also, an increase in the temperature of the nuclear fuel may result in the generation of hydrogen. Such circumstances have the potential to exert a considerable influence on the structural integrity of the containment. In South Korea, the Containment Filtered Venting System (CFVS) has been identified as a potential mitigating measure for severe accidents. The CFVS is a pressure stability system that facilitates the venting of atmospheric gases from the containment to the atmosphere [1]. However, there is still the problem of the ultimate release of radioactive material to the environment, even though the CFVS is equipped with a decontamination system designed to remove as much radioactive material as possible. In South Korea, it is inappropriate to release radioactive materials generated inside the containment during an accident, due to legislative changes following the enactment of severe accident regulations. Therefore, the application of CFVS is not feasible in South Korea [2].

Therefore, we aim to conduct foundational research on a passive airborne radioactive material reduction system that uses the catalytic reaction of hydrogen to reduce radioactive materials without releasing radioactive materials into the environment. The catalyst-filled bed utilized in the system comprises spherical particles of an alumina-based ion-exchanged material impregnated with Pt, which serves to catalyze the reaction between hydrogen and oxygen, thereby producing vapor and resulting in an exothermic reaction [3]. The exothermic reaction of the catalysts increases the temperature of the air within the housing, creating an upward flow that induces natural convection within the system. The natural circulation effect induces a continuous flow of air from the containment building into and out of the system, thereby sustaining this process until a designated amount of hydrogen is consumed. Within the system, filters and absorbers are installed for the purpose of ensuring the continuous removal of radioactive aerosols and gases from the containment atmosphere during the natural circulation process. The most important part of these systems is the formation of natural convection and the evaluation of the performance of the catalysts installed inside the passive filtration system.

In this paper, an experimental apparatus was constructed to investigate the exothermic properties of the catalyst in relation to hydrogen concentration and flow rate. The data obtained served as a basis for determining essential information and design variables for passive airborne radioactive material reduction system.

2. Description of Experimental Apparatus

The generation of passive flow using an exothermic catalyst is critical to the effectiveness and operation of a passive filtration system. To evaluate the exothermic properties of a catalyst-filled bed under controlled inlet conditions, an Apparatus of Catalyst-filled Bed Characteristics Evaluation (ACE) has been developed. Although the actual system operates under natural convection, it is important to obtain quantitative data on the exothermic characteristics of the catalyst-filled bed to support. Forced convection experiments were therefore carried out to better control the inlet fluid conditions such as the mass flow rate and the hydrogen concentration.

2.1 Catalytic Material

Since the catalyst support material is required to operate under severe accident conditions in nuclear power plants, it must exhibit structural integrity in high temperature and high humidity environments. To address this issue, activated alumina (α-Al_2O_3) was selected as an inorganic porous material due to its exceptional heat resistance and moisture resistance. Furthermore, this material was then ion-exchanged with 0.5 wt% Pt to create an exothermic catalyst designed for use with combustible gases.

2.2 Experimental setup

As shown in Figure 1, the ACE system comprises several major components: a lower disc-shaped buffer tank, inlet duct, catalytic bed, gas sampling line and exhaust duct. Gases (such as air and hydrogen) are injected into the buffer tank through two symmetrical nozzles. The flow rate of gases into the buffer tank is controlled by the MFC (Mass Flow Controller). To reduce the jet effect, a sintered metal filter is placed at the end of the nozzle. In addition, a porous plate is placed on top of the buffer tank to facilitate gas mixing. A hydrogen and oxygen concentration sampling manifold is located at both the front and rear of the catalyst-filled bed to allow measurement of hydrogen and oxygen consumption within the catalytic bed. A condenser was installed upstream of the gas sampling to eliminate moisture, along with a rotameter to ensure a stable gas flow rate. In this experiment, the steam production from the catalytic reaction of hydrogen was not measured separately because the equipment was designed to determine the exothermic properties of the catalyst according to the inlet conditions (gas concentrations, velocity) of the experimental apparatus.

The catalyst-filled bed section contains two vertical catalyst beds (dimensions: width 5 mm, length 100 mm, height 150 mm), each containing 53 g of spherical catalyst packed in the bed. Figure 2 shows the placement of thermocouples (TCs) in the flow-induced section, which are used to calculate the heat rate of the catalyst beds. To reduce heat loss, the gas injection line, buffer tank and flow-induced section are insulated. The average error in the heat rate measured during the experiment was approximately 1.6%.

The evaluation of the exothermic characteristics of the catalyst-filled bed was carried out under forced convection conditions with varying hydrogen concentrations and flow rates. The tests were carried out at a concentration of 8 vol% to avoid the risk of combustion, which indicated that combustion could occur at hydrogen concentrations of around 9%. The total gas flow rate was adjusted within a range of 60 to 190 LPM. The test matrix for dry conditions is shown in Table I.

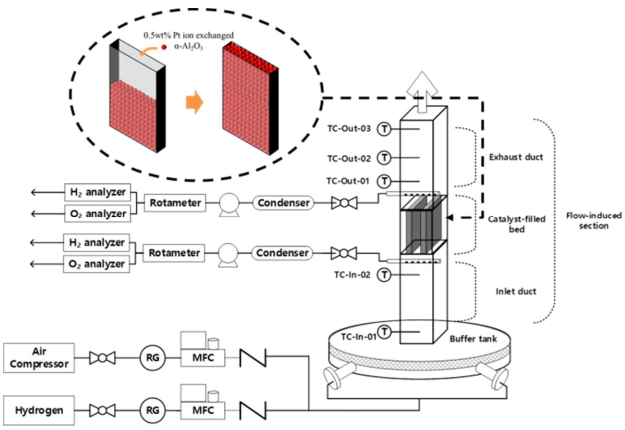

Figure 1. Schematic of experimental setup

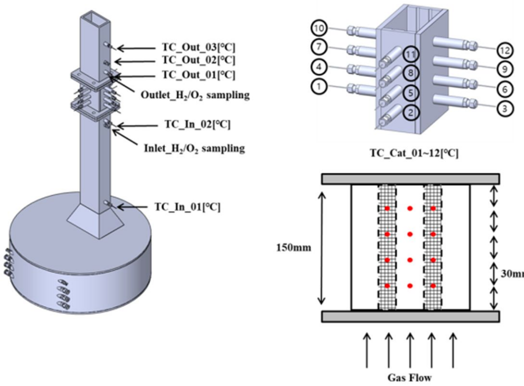

Figure 2. The experimental facility and measurement points

Table I: Summary of test matrix under the dry conditions

Test Cases	H_2 [vol %]	Air [vol %]	O_2 [vol %]	Hydrogen Velocity, [m/s]	Total Volume Flow, [LPM]	Initial Temperature, [°C]
1	2.1	97.9	19.5	0.2	60	25
2	2.7	97.3	19.7			
3	4.3	95.7	19.6			
4	7.1	92.9	21.1			
5	1.1	98.9	20.9	0.4	126	
6	1.3	98.7	20.9			
7	2.1	97.9	20.8			
8	2.8	97.2	20.6			
9	3.6	96.4	20.5			
10	4.4	95.6	20.3			
11	7.9	92.1	20.8			
12	0.8	99.2	22.4	0.6	190	
13	1.0	99.0	22.3			
14	1.5	98.5	22.2			
15	2.1	97.9	22.2			
16	2.7	97.3	22.0			
17	3.2	96.8	21.9			
18	3.8	96.2	21.9			
19	4.4	95.6	20.6			
20	8.3	91.7	20.9			

3. Results and Discussion

Figure 3 shows the temperature difference between the gas in front of the catalyst-filled bed and the gas between the two catalytic beds as a function of hydrogen velocity. Temperature variation increases linearly with hydrogen volume concentration, while velocity has negligible effect on temperature rise at constant hydrogen concentration. This indicates that at lower velocities, hydrogen entry into the catalyst-filled bed is reduced, increasing the residence time and leading to a higher hydrogen recombination rate.

In Figure 4, to show the difference in gas temperature in the catalytic bed region, the volume fraction of hydrogen supplied is converted to mass flow rate. In contrast to Figure 3, and confirming that this result is related to the rate of hydrogen removal as it depends on flow rate, the difference in exothermic temperature compared to mass flow rate tends to increase sharply at lower flow rates.

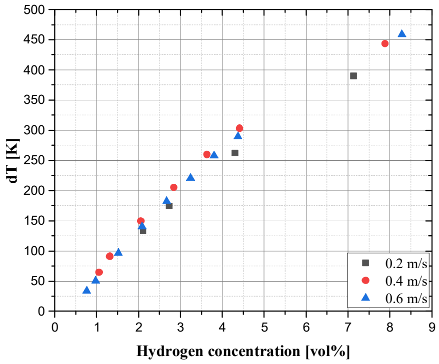

Figure 3. Gas temperature changes with hydrogen concentration

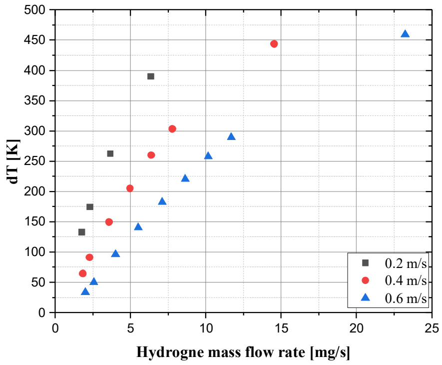

Figure 4. Gas temperature changes according to hydrogen mass flow rate

As shown Figure 5, the heat rate of the catalytic bed was determined from the volume concentration of hydrogen. The heat produced by the catalytic bed is controlled by the stoichiometric reaction ratio between hydrogen and oxygen. As a result, an increase in the hydrogen velocity results in a greater supply of mass for hydrogen and oxygen, which in turn causes the heat rate to increase.

It was confirmed that the concentration and velocity of hydrogen and oxygen are the principal factors affecting the heat generated by the catalyst. Based on this, a correlation was developed, as illustrated in Equation 1, to predict the heat generated by the catalyst under varying inlet conditions. As shown in Figure 6, this correlation is an accurate prediction of the experimental data within a 20% margin.

$$Q = 2200 + \frac{v^{0.827} \cdot {C_{H_2}}^{0.7}}{{C_{O_2}}^{1.3}} \qquad (1)$$

Where, Q [w] is the heat rate of catalyst-filled bed, v is the inlet velocity of experimental facility [m/s] and C is the volume concentration [%]. The empirical correlation predicts the exothermal heat rate of a Pt-coated αAl_2O_3 Catalyst in Bed type and is applicable in dry conditions with hydrogen concentration of 10 % or less.

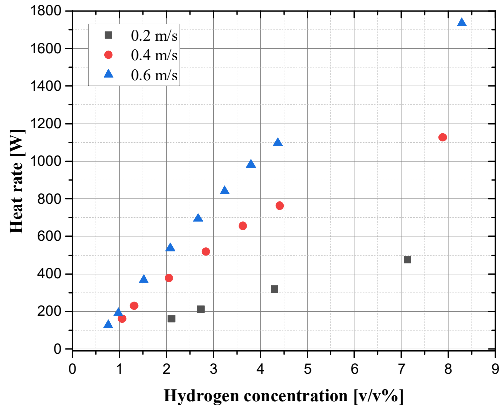

Figure 5. Heat rate of the catalyst-filled bed with hydrogen concentration

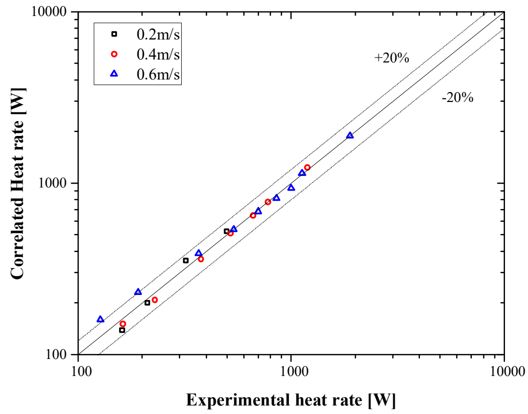

Figure 6. Comparison between the experimental data and the correlation data

4. Conclusions and Future Works

In this paper, the concept of a passive radioactive material removal system was presented and the experimental facility was constructed to evaluate the exothermic properties of the catalyst.

In the tests, the exothermic properties of the catalyst have been evaluated under various inlet conditions (gas flow rate and volume concentration). Based on the evaluation of the exothermic properties of the catalyst, an experimental correlation was presented to predict the heat rate of the catalyst-filled beds. The experimental correlation was found to predict the experimental results very well, within 20%.

In addition, the developed experimental facility will be used to evaluate the exothermic properties of the catalyst under conditions that include humidity, reflecting the atmosphere inside the containment building. Finally, the data obtained will also be used to design the catalyst for a real passive airborne radioactive material reduction system.

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