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PAR Auto-ignition and Its Effect on Hydrogen Mitigation of a Pressurized Water Reactor

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1. Introduction

Currently, PAR (passive autocatalytic recombiner) and hydrogen igniters are mainly used around the world to mitigate hydrogen in pressurized water reactor (PWR) containments in the event of severe accidents [1, 2]. PAR and hydrogen igniter, which are installed in containment buildings for the purpose of preventing hydrogen combustion accompanied by strong shock waves such as detonations, have different characteristics. While PAR operates (recombine hydrogen) when hydrogen comes into contact with the PAR catalyst without operator intervention, the hydrogen removal rate is very low. If only PAR is installed in the containment building, the distribution and removal characteristics of hydrogen in the event of a severe accident differ depending on the thermal-hydraulic characteristics of the reactor and the containment building and the accident progression. Hydrogen ignitors and PARs are installed in the containment buildings of APR1400 [3]. A hydrogen igniter can quickly remove hydrogen, but requires intervention such as an external power source and operator judgment and operation. In the event of a station blackout (SBO) accident, the operation is impossible due to lack of electric power supply, so there is a trend to install batteries or uninterruptible power supply (UPS) as auxiliary power sources (AP1000 [4]).

A hydrogen igniter can be installed around the hydrogen release point to burn and eliminate hydrogen in the form of a diffusion flame, but it is important to ensure the integrity of surrounding equipment from the heat generated by the continuous combustion flame. As an active device, the timing of operation of hydrogen igniter is very important. If an accident in a PWR plant progresses into a severe accident, it is appropriate to operate the hydrogen igniter immediately. However, if the operation of the hydrogen igniter is delayed for various reasons, that is, after a large amount of hydrogen has already been released into the containment building, the operation of the hydrogen igniter becomes an important issue.

While the passive characteristic of PAR is a very effective advantage compared to hydrogen igniter, it has the disadvantage of delayed operation of PAR. In PAR, when the catalytic chemical reaction of hydrogen that reaches the surface of the catalyst body through diffusion occurs initially, the temperature of the catalyst body rises and high-temperature exhaust gas is emitted. As a result, new hydrogen mixed gas flows into the PAR inlet, thereby increasing the PAR recombination. The catalytic reaction continues. The temperature rise of the PAR catalyst is closely related to the hydrogen removal rate and hydrogen combustion by PAR. When the temperature of the PAR catalyst rises above about 1,000 K, the catalyst acts as an ignition source of hydrogen, and this phenomenon is called PAR-induced combustion [5, 6] or PAR auto-ignition [7]. Auto-ignition of PAR, along with initial operation delay, is a representative characteristic of PAR and a representative issue in PAR-based hydrogen mitigation of PWRs.

The recombination load on PAR is determined by the hydrogen concentration flowing into the inlet. Hydrogen concentration below 2 % affects the initial start-up delay of PAR, regular hydrogen recombination occurs around 4 %, and above 6 %, PAR is overloaded. It is understood that an increase in the temperature of the overloaded catalyst causes spontaneous ignition of hydrogen.

To date, the detailed mechanism of PAR auto-ignition has not been revealed, but it is understood that hydrogen flame generated by PAR mainly spreads upward through the PAR housing.

In this study, we analyze the experimental results of PAR auto-ignition and discuss the impact on hydrogen mitigation for an accident management.

2. Analysis of Experimental Data

In the THAI-1 project [8], 30 experiments (HR tests) were performed under various thermal hydraulic conditions using AREVA, NIS, and AECL PAR. Of these, PAR-induced combustion occurred in the 22 test cases and it n did not occur only in 8 test cases. The THAI-1 HR experiment procedure is divided into a preconditioning stage to form the initial thermal hydraulic conditions in the test vessel and a hydrogen injection stage. The hydrogen injection rate is between 0.1 and 0.5 g/s. If the hydrogen injection rate is initially started low, it increases by about two times after some time. After a certain period of time, hydrogen injection is stopped, and when the hydrogen concentration at the PAR inlet continues to decrease to below 1%, the second stage hydrogen injection is performed. In the THAI-1 HR experiment, concentration was measured using 15 hydrogen sensors and 2 oxygen sensors, and gas temperature, pressure, and wall temperature were also measured.

Fig. 1 is the result of the THAI HR-1 experiment using AREVA PAR, showing the hydrogen flow rate injected into the experimental vessel over time and the hydrogen removal rate according to the hydrogen concentration at the PAR inlet.

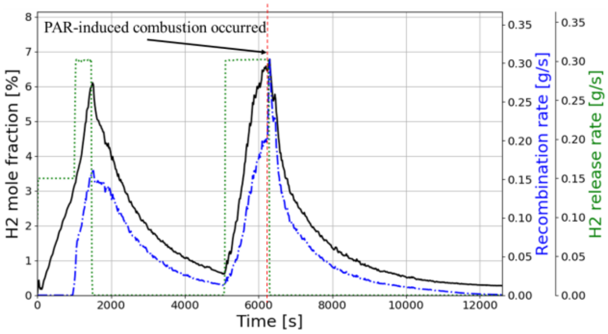

Fig. 1. THAI HR-1 test results of PAR inlet hydrogen mole fraction, hydrogen release rate, and recombination rate over time.

We aim to establish the requirements for PAR auto-ignition by analyzing the thermal hydraulic conditions for each experiment, especially the conditions at the time of PAR-induced ignition. Representative factors that can predict the occurrence of spontaneous combustion due to PAR during an experiment include gas pressure, temperature, species concentrations and catalyst temperature, and indirectly, hydrogen combustion can be predicted through a large reduction rate in hydrogen concentration.

In order to analyze the spontaneous combustion of PAR, it is necessary to distinguish the major thermal hydraulic factors mentioned above into cause and effect. The increase in pressure and gas temperature inside the test vessel is a typical result of spontaneous ignition. The large decrease in hydrogen concentration in the vessel is also a result of spontaneous combustion. On the other hand, the catalyst temperature and hydrogen concentration at the PAR inlet are representative causes of spontaneous ignition of PAR.

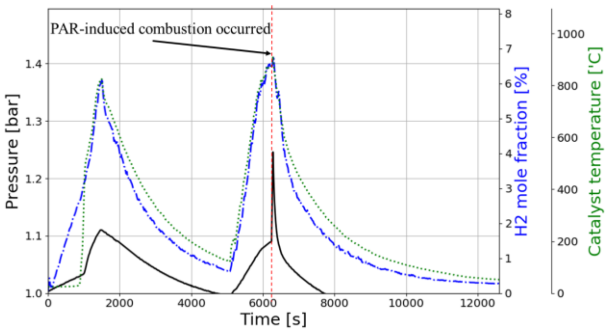

Fig. 2. THAI HR-1 test results of test vessel pressure, PAR inlet hydrogen mole fraction, and catalyst temperature over time.

Fig. 2 shows changes in pressure, PAR inlet hydrogen concentration, and catalyst temperature over time in the THAI HR-1 test. When spontaneous ignition by PAR occurs, a rapid increase in pressure can be seen, and at this time, the temperature of the catalyst and the hydrogen concentration at the PAR inlet are the main causes of spontaneous ignition by PAR.

Spontaneous ignition and hydrogen combustion by PAR are abnormal phenomena in which the pressure wave and flame front propagate after ignition, and it is difficult to accurately predict the time of spontaneous ignition because it occurs in a very short time and is also related to the response speed of the measurement sensor. Here, the timing of PAR auto-ignition was quantified based on the rapid change in pressure within the test vessel.

As shown in Fig. 2, it can be seen through the pressure change in the test vessel that combustion induced by PAR occurred 6260 seconds after the start of hydrogen injection in the HR-1 experiment. At 1480 seconds, when the first hydrogen injection ended, the hydrogen concentration at the PAR inlet rose to 6%, but spontaneous ignition did not occur. At 6260 seconds, when the second hydrogen injection ended, the hydrogen concentration at the PAR inlet reached 6.5% and then spontaneous ignition occurred. At this time, it can be seen that the temperature of the PAR catalyst is maximum.

Table 1 quantitatively evaluates the concentrations of hydrogen and oxygen at the PAR inlet, the hydrogen concentration at the outlet, and the average and maximum temperature of the catalyst at spontaneous ignition for the test cases in which PAR auto-ignition occurred among the HR tests of THAI-1.

Table 1. THAI-1 HR test cases with ignition occurring, conditions at PAR auto-ignition

Test	Vendor	t_{Auto}	H_{2In}	H_{2Out}	O_{2In}	Tc_{Avg}	Tc_{Max}
HR-1	AREVA	6260	6.50	2.4	17.3	891	892
HR-2	AREVA	5580	6.90	2.5	17.8	894	894
HR-3	AREVA	5500	6.37	2.8	17.6	889	889
HR-4	AREVA	2180	6.59	2.8	18.5	712	760
HR-6	AREVA	1740	7.51	2.8	13.6	903	962
HR-7	AREVA	10320	7.28	2.7	13.2	899	950
HR-8	AREVA	2040	7.60	2.7	13.5	907	969
HR-10	AREVA	1280	8.04	2.8	9.3	916	966
HR-15	NIS	7500	6.35	2.7	11.8	670	755
HR-16	NIS	1980	7.04	3.1	13.6	692	773
HR-17	AECL	6960	6.99	4.3	17.6	704	755
HR-18	AECL	6900	6.85	4.2	17.8	700	759
HR-19	AECL	8900	8.36	4.9	14.0	737	785
HR-20	AECL	9380	8.34	4.7	11.7	753	789
HR-23	AECL	6460	7.40	4.4	18.7	701	748
HR-24	AECL	9080	7.27	4.7	18.2	712	743
HR-25	AECL	9040	7.11	4.7	19.0	699	720
HR-26	AECL	9280	6.88	4.7	18.1	707	744
HR-27	AREVA	7120	6.50	2.8	17.9	836	895
HR-28	AREVA	7080	6.35	2.9	17.8	844	897
HR-29	AREVA	4760	8.77	2.9	9.8	921	966
HR-30	AREVA	5100	8.16	3.7	9.5	972	1017

Based on Table 1, the conditions for spontaneous ignition of each PAR are summarized as follows.

AREVA PAR: $x_{H2} > 6.35$ %, $T_{cat} > 760$ ℃ (1)
NIS PAR : $x_{H2} > 6.35$ %, $T_{cat} > 755$ ℃ (2)
AECL PAR : $x_{H2} > 6.85$ %, $T_{cat} > 720$ ℃ (3)

Spontaneous ignition of PAR is also affected by the oxygen concentration of the gas flowing into the PAR inlet, and in experiments where the oxygen concentration was 10% or less, spontaneous ignition occurred when the hydrogen concentration was 8% or more.

Among the 30 cases performed in the THAI-1 HR experiment, the cases in which spontaneous ignition did not occur are shown in Table 2. In HR-5, HR-12, and HR-14, it appears that spontaneous ignition did not occur because the hydrogen concentration at the PAR inlet did not meet the hydrogen concentration conditions of equations (1)-(3), and in other cases (HR-9 , HR-11, HR-13, HR-21, and HR-22), it appears that spontaneous ignition did not occur due to low oxygen concentration. In the case of HR-9, the hydrogen concentration at the PAR inlet reached 9.4%, but ignition by PAR did not occur. This is related to the fact that under oxygen starved conditions the hydrogen concentration participating in PAR hydrogen recombination is equal to the oxygen concentration. In other words, the hydrogen concentration equivalent to the oxygen concentration of 6.3% did not satisfy equation (1), and therefore spontaneous ignition did not occur.

Table 2. THAI-1 HR test cases without ignition occurring, conditions at maximum hydrogen concentration at PAR inlet.

Test	Vendor	t_{H2Max}	H_{2In}	H_{2Out}	O_{2In}	Tc_{Avg}	Tc_{Max}
HR-5	AREVA	2080	5.6	3.1	19.7	841	844
HR-9	AREVA	8800	9.4	5.1	6.3	745	756
HR-11	AREVA	6460	7.0	4.8	3.4	484	498
HR-12	AREVA	8440	6.1	4.1	4.2	520	535
HR-13	AREVA	6500	6.8	4.7	2.7	424	437
HR-14	NIS	11700	6.1	4.4	3.2	363	397
HR-21	AECL	1880	7.4	6.9	1.3	225	234
HR-22	AECL	7980	7.0	5.6	3.4	419	434

The way PAR is loaded can vary depending on the distribution of hydrogen flowing into the installed compartment. In other words, when a high concentration of hydrogen gas mixture is introduced, the PAR immediately enters an overload state and spontaneous combustion occurs. When the hydrogen concentration gradually increases from lean condition, high-temperature exhaust gas is continuously discharged above the PAR due to regular hydrogen recombination. Even if the PAR is overloaded and spontaneous combustion occurs due to an increase in hydrogen concentration, the hydrogen combustion pressure can be relatively lowered because the hydrogen concentration in the space above the PAR is low.

In Table 3, pressure increase by PAR auto-ignition in the HR tests is compared to the AICC (adiabatic isochoric complete combustion) pressure. The AICC pressure is calculated based on the conditions before the auto-ignition. In the table, AICC ration is defined as follows:

$$\text{AICC ratio: } \frac{p_{Max}-p_{ign}}{p_{AICC}-p_{ign}} \quad (4)$$

Table 3. THAI-1 HR test cases with ignition occurring, comparison of auto-ignition pressure and AICC pressure.

Test	p_{Ign}	p_{Max}	p_{AICC}	AICC ratio
HR-1	1.17	1.25	2.94	0.04
HR-2	1.22	1.24	3.01	0.01
HR-3	1.68	1.78	4.02	0.04
HR-4	2.87	2.92	6.17	0.01
HR-6	2.02	2.32	4.95	0.10
HR-7	1.54	1.80	3.74	0.12
HR-8	1.73	1.93	4.18	0.08
HR-10	1.79	2.33	4.40	0.21
HR-15	1.60	1.60	3.32	0.00
HR-16	1.67	1.74	3.66	0.03
HR-17	1.76	2.13	4.53	0.13
HR-18	1.65	2.17	4.31	0.19
HR-19	1.53	1.84	3.73	0.14
HR-20	1.67	2.02	4.03	0.15
HR-23	1.31	1.64	3.48	0.15
HR-24	2.90	2.97	6.83	0.02
HR-25	2.91	2.98	6.86	0.02
HR-26	2.69	2.83	6.49	0.04
HR-27	1.75	1.83	4.39	0.03
HR-28	1.65	1.78	4.04	0.06
HR-29	1.64	2.06	4.17	0.17
HR-30	3.10	3.65	8.02	0.11

Typically, the pressure rise caused by hydrogen combustion in a closed compartment with uniform distribution of hydrogen concentration is more than 50 % of the AICC pressure. Table 3 depicts that the AICC ratio is below 20 % in all the cases of THAI-1 HR tests with combustion occurring. It means that the pressure build-up by PAR auto-ignition is weak.

3. Effect of PAR auto-ignition on Hydrogen Mitigation

In the event of a severe accident, hydrogen mitigation in the PWR containment building is to prevent shock wave combustion (detonation) in the containment building. Detonation occurs through several continuous stages: ignition, flame acceleration (FA), and deflagration-to-detonation transition (DDT). In other words, even if there is an ignition source, detonation cannot occur if there is no flame acceleration. DDT occurs when a high-speed turbulent flame front catches up a preceding shock wave, and through many DDT experiments [], it is known that there has been no case where DDT occurred when the concentration of hydrogen mixed with air was less than 10% [9]. It depicts that DDT does not occur when the high-speed, turbulent flame, which is a pre-requisite condition for DDT, has a flame acceleration (FA) index less than 1. From a thermodynamic point of view, the FA index is less than 1 when the hydrogen concentration is less than 10%.

The hydrogen concentration, which is the condition for PAR auto-ignition, is about 7%, which is much lower than the hydrogen concentration for DDT. Equation (5) represents the correlation among flammability conditions, auto-ignition conditions, and DDT conditions.

$$x_{H2,flammable} < x_{H2,auto-ignition} < x_{H2,DDT} \qquad (5)$$

In other words, since the hydrogen concentration of auto-ignition is lower than that of DDT, it indicates that auto-ignition cannot be the direct cause of DDT.

There are two main mechanisms by which shock wave combustion, or detonation, occurs inside a containment building composed of many compartments. The first is that the flame accelerates as the flame surface wrinkles and stretches while passing through compartments or corridors with obstacles [9], causing DDT to break out. Second, DDT can occur in a duct or channel-type flow path that is much longer than the hydraulic diameter due to the interaction between the wall boundary layer of a flow and the flame front and the overlap of pressure waves [10]. Therefore, when PAR acts as an ignition source in a complex-shaped containment building, DDT cannot be ruled out if the path of movement of the flame is very long and there are many obstacles. However, even in this case, it is assumed that a hydrogen cloud with an FA index greater than 1 has been formed. Therefore, in order to prevent the occurrence of DDT due to auto-ignition of PAR, the arrangement of PARs according to the shape of the containment building and the characteristics of the nuclear reactor is very important.

4. Conclusions

As the concentration of hydrogen flowing into PAR increases, the catalyst recombination rate increases and the catalyst temperature rises, causing auto-ignition, and this phenomenon can be seen as one of the characteristics of PAR.

In particular, the hydrogen concentration threshold for PAR auto-ignition may vary depending on PAR, but is lower than the hydrogen concentration that can generate DDT. In a containment building where hydrogen is distributed, auto-ignition by PAR can generate a hydrogen flame at lower concentration of hydrogen than that of DDT. Therefore, a gas mixture with a hydrogen concentration capable of generating DDT may be burned by spontaneous ignition of PAR before it is formed. However, when the hydrogen flame generated by PAR meets a highly concentrated hydrogen cloud, the possibility that it transitions into a fast turbulent flame or DDT cannot be ruled out. Therefore, PAR installation (locations and number of PARs to be installed) can be a significant issue.

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