The Study on Permissible Value of Hydrogen Gas Concentration in Purge Gas of Fuel Cell Vehicles

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ABSTRACT
Ignition conditions and risks of ignition on a permissible value of hydrogen concentration in purge gas prescribed by HFCV-GTR were reevaluated. Experiments were conducted to investigate burning behavior and thermal influence of continuous evacuation of hydrogen under continuous purge of air / hydrogen premixed gas, which is close to an actual purge condition of FCV, and thermal evacuation of hydrogen. As a result of the reevaluation, it was shown from the viewpoint of safety that the permissible value of hydrogen concentration in purge gas prescribed by the current HFCV GTR is appropriate.

1. INTRODUCTION

For fuel cell vehicles (FCV), hydrogen gas purging is undertaken in order to discharge into general surroundings the impurities-containing hydrogen gas accumulated inside FCV fuel cell systems. To ensure the safety of hydrogen gas purging, the UN regulation HFCV-GTR Phase 1 (GTR No.13)[1] published in June 2013 specifies the maximum allowable hydrogen concentration of purged gas to be 4vol% on 3-second average and 8vol% at any time. These safety criteria were based on the results of ignition simulation tests[2] carried out under the condition of air/hydrogen premixed gas continuously flowing out from an exhaust pipe. Nevertheless, the actual conditions for purging hydrogen from the FCV fuel cell system differ from the theoretical purging conditions applied to simulation tests. For example, while simulation assumes purging of fuel cell systems with air taken from the atmosphere, the actual purge gas differs from atmospheric air in containing less oxygen but more water vapor because the polymer electrolyte fuel cells consume oxygen in purge gas. Furthermore, while simulation assumes continuous and steady discharge of air/hydrogen premixed gas for purging, actual purging involves sporadic mixing of hydrogen into a continuous stream of air. Consequently, simulated and actual purged gases differ not only in their oxygen concentration and humidity levels but also in their lower flammable limit, flame holding limit, various other combustion characteristics, and ignition risks around the FCV exhaust outlet. The present study was therefore conducted to reevaluate the ignition conditions and associated risks of purged gas in relation to the maximum allowable hydrogen concentration set down in HFCV-GTR Phase 1. First, combustion behavior was investigated under the condition of continuous purging with air/hydrogen premixed gas in order to determine the levels of lower flammable limit, flame holding limit, and flashback limit when the air flow rate, oxygen concentration, and humidity of purged gas were varied. Second, combustion behavior, thermal influence around the exhaust outlet, and any injury to nearby persons were examined under the hydrogen concentration condition prescribed in HFCV-GRT Phase 1 and also under the condition of sporadic mixing of hydrogen into a continuous stream of discharged air.

2. COMBUSTION BEHAVIOR UNDER CONTINUOUS DISCHARGE OF AIR/HYDROGEN PREMIXED GAS

In the present study, combustion behavior under the continuous discharge of air/hydrogen premixed gas was investigated while premixed gas concentration, discharge flow rate, oxygen concentration, and humidity were set as parameters.

2.1. Test method

Figure 1 shows the exhaust pipe model employed in the present tests. Air and hydrogen stored separately in cylinders were released into two separate 1/2-inch pipes; then, made to mix together in a single 1-inch pipe; finally discharged into surroundings through a 2-inch (52 mm inner diameter) straight pipe consisting of a 2,000 mm long polycarbonate pipe and a 250 mm long SUS stainless steel pipe for determining flashback possibility. The final straight pipe had an inner diameter practically identical with that of the exhaust pipes fitted on FCVs currently on sale. The gas flow rates were controlled with mass flow controllers (MFC). The concentration of air/hydrogen premixed gas was calculated from the ratio of MFC-controlled flow rates of air and hydrogen gas, and was confirmed to be equal to the premixed gas concentration measured prior to the testing. Three different air flow rates of 50, 500, and 2,000 NL/min were applied to cover the whole flow rate range of purged gas for commercialized FCVs. Two oxygen concentration values of 10.0vol% and 20.8vol% (the latter equal to the atmospheric oxygen concentration) and two humidity values of 0RH% and 99RH% were also applied. In the relative humidity 99RH% test, hot water placed in a 300L bubbling chamber was heated to approximately 85°C, and air was made to pass inside the chamber to be moistened with the steam.

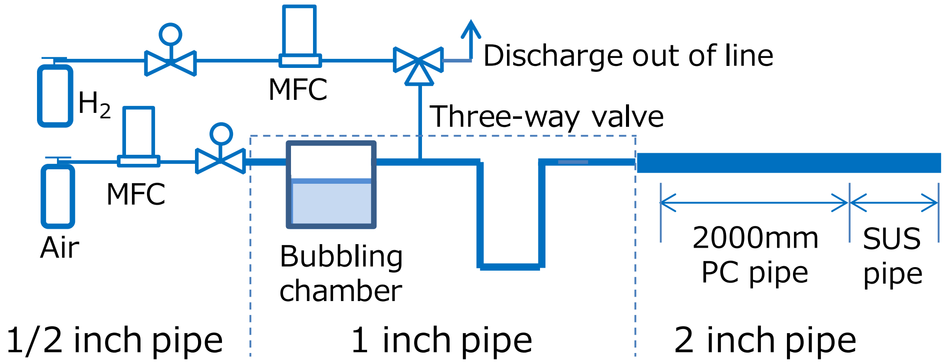

Figure 1. Schematic of exhaust pipe model with bubbling chamber

For ignition operation, air/hydrogen premixed gas was released for over 1 minute until its flow rate and hydrogen concentration became stable. Then, ignition was tested by applying direct-current discharge of 3 mm gap length and a 30 mJ ignition energy to the brim center of the FCV exhaust outlet. When an ignition phenomenon was detected, the DC discharge was stopped and the combustion behavior of air/hydrogen premixed gas was examined. Not visually recognizable in most cases, the occurrence or non-occurrence of ignition was judged from images taken by an infrared thermal imaging camera. The ignition test was conducted in an indoor experiment facility (diameter 18m, height 16m) free from the influence of the wind and weather.

2.2. Test results and discussion

Figures 2 and 3 show ignition behavior examples captured by the infrared thermal imaging camera under the test condition of a 50 NL/min air flow rate and hydrogen concentrations of 8.6vol% and 4.6vol%.

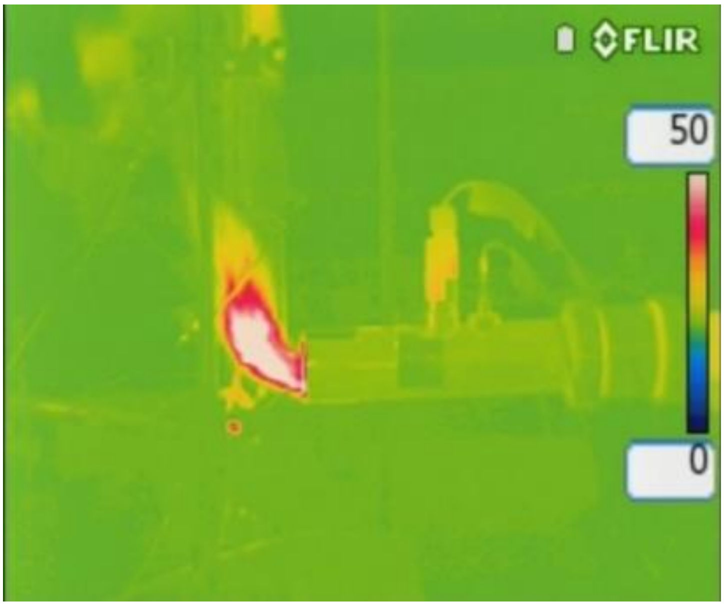

Figure 2. Ignition behavior (8.6Vol.%, Air 50NL/min)

Figure 3. Ignition behavior (4.6Vol%, Air 50NL/min)

The combution clearly proved to have occurred when the hydrogen concentration was 8.6vol%. On the other hand, the combution of at a 4.6vol% hydrogen concentraion was very small. However, at the same 4.6vol% hydrogen concentration, flame could be maintained continuously even after the DC discharge had been ceased. It was found that at a lower 4.2vol% hydrogen, flame extinguished itself after DC discharge was stopped. In contrast, at 8.6vol% hydrogen, combustion took place outside the exhaust pipe immediately after ignition, and flashback was generated gradually inside the exhaust pipe.

The judgment of combustion was made from such a combustion image of the infrared thermal imaging camera. The results of the ignition test of the air / hydrogen premixed gas with the air flow rate as a parameter are shown in Figure 4 and lower flammable limit (LFL), flame holding limit (FHL), and flashback limit are shown in Table 1.

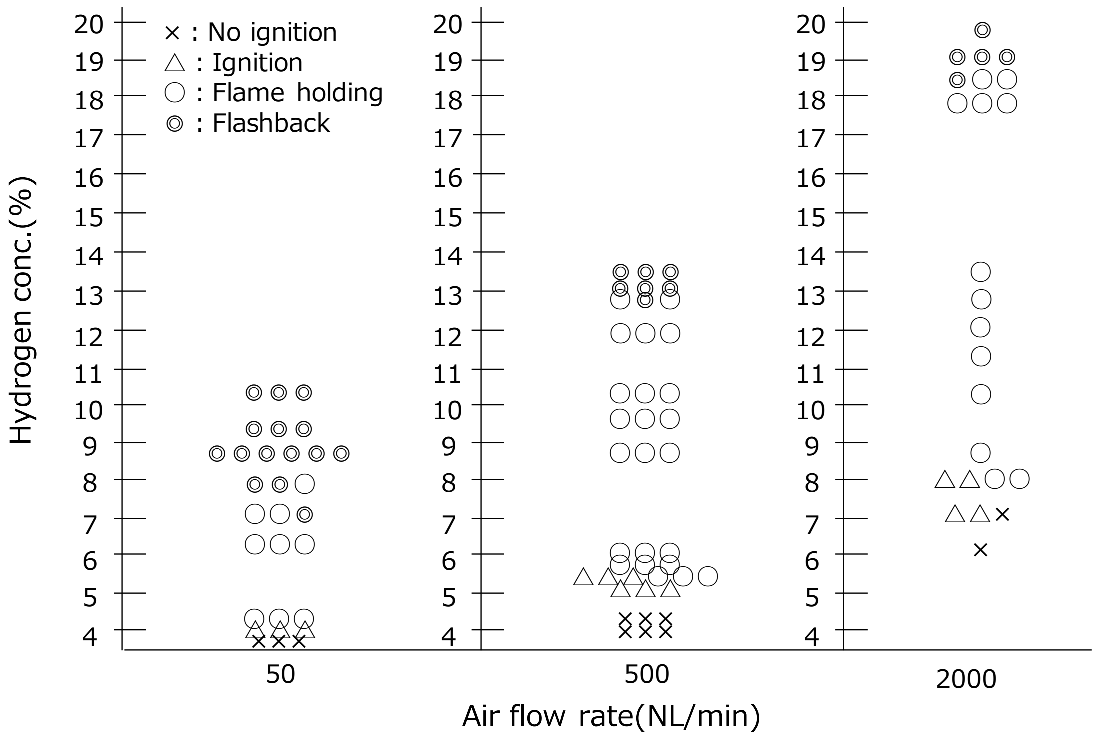

Figure 4. Result of the ignition tests with air flow rate
(Oxygen conc.20.8 Vol.%, Humidity 0 RH%)

Table 1. Result of LFL, FHL, and flashback limit with air flow rate

Air Flow (NL/min)	O_2 Conc. (Vol.%)	Relative humidity (RH%)	Hydrogen Concentration (Vol.%)		
			Lower Flammable Limit	Flame Holding Limit	Flashback Limit
50	20.8	0	4.2	4.6	8.8
500			5.3	5.8	13.3
2000			7.4	9.1	19.4

As evident in the above figure, the LFL, FHL and flashback limit all required a higher hydrogen concentration as the air flow rate was increased. An air flow rate of 50 NL/min (and an air/hydrogen premixed gas speed of 0.4 m/s) resulted in a 4.2vol% LFL, a 4.6vol% FHL, and 8.8vol% flashback limit, which were all comparable with the test results given in the referenced studies[3],[4] by other researchers.

The study[2] which had been referenced to determine the maximum allowable hydrogen concentration in HFCV-GTR Phase 1 reported that a hydrogen concentration of 8vol% or higher was necessary for flame holding to take place. Yet the present study's FHL values of 4.6vol% and 5.8vol% hydrogen at air flow rates of 50 NL/min and 500 NL/min respectively indicated the occurrence of flame holding at hydrogen concentration lower than the 8vol% reported in the referenced study.

The results of the ignition test of the air / hydrogen premixed gas with the oxygen concentration and humidity as a parameter are shown in Figure 5 and lower flammable limit (LFL), flame holding limit (FHL), and flashback limit are shown in Table 2.

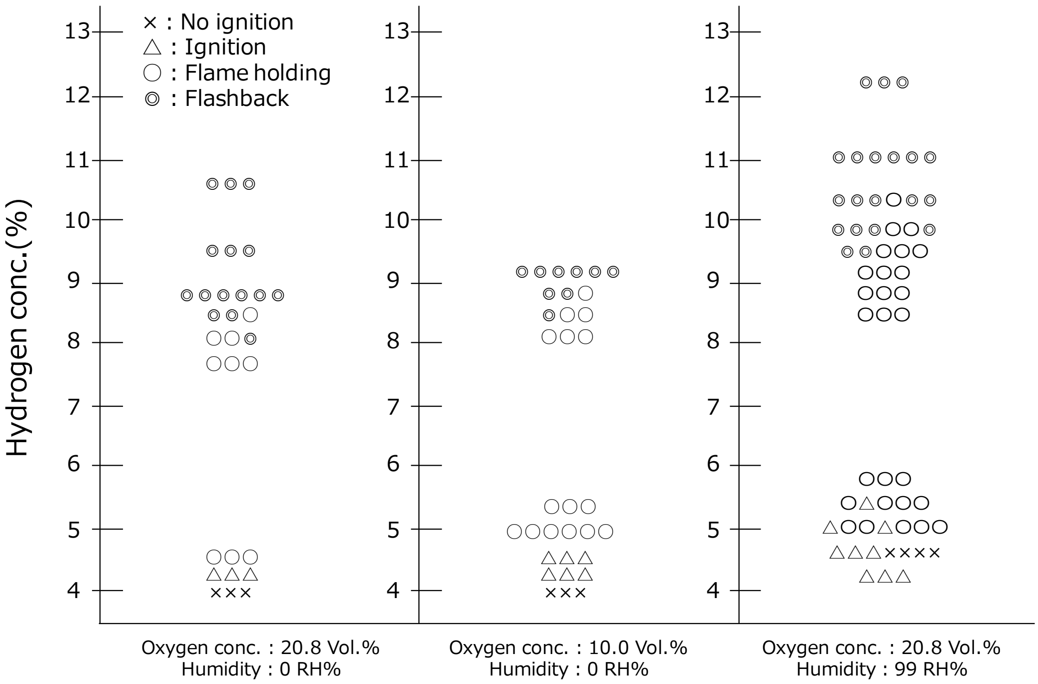

Figure 5. Result of the ignition tests with oxygen concentration and humidity
(Air flow rate : 50NL/min)

Table 2. Result of LFL, FHL, and flashback limit with oxygen concentration and humidity

Air Flow (NL/min)	O_2 Conc. (Vol.%)	Relative humidity (RH%)	Hydrogen Concentration (Vol.%)		
			Lower Flammable Limit	Flame Holding Limit	Flashback Limit
50	20.8	0	4.2	4.6	8.8
50	10.0		4.2	4.9	9.1
50	20.8	99	4.6	5.7	9.7

Where test results were compared between oxygen concentrations of 10.0vol% and 20.8vol%, the LFL proved identical between the two oxygen concentrations but the FHL rose from 4.6 to 4.9vol% and the flashback limit from 8.8 to 9.1vol%, indicating a maximum 0.4vol% shift toward higher hydrogen concentration. Similarly, where test results were compared between 0RH% and 99RH%, the LFL rose from 4.2 to 4.6vol%, the FHL from 4.6 to 5.7vol%, and the flashback limit from 8.8 to 9.7vol%, indicating a maximum 1.1vol% shift toward higher hydrogen concentration. Nevertheless, these changes proved smaller than the hydrogen concentration changes shown in Figure 4, so that the lower flammable limit, the flame holding limit, and the flashback limit were found more dependent on purged gas flow rates than on its oxygen concentration and humidity factors.

3. COMBUSTION BEHAVIOR UNDER HFCV-GTR-PRESCRIBED HYDROGEN CONCENTRATION

In the next test, hydrogen gas was released sporadically into a continuous stream of air under conditions approximating the purged gas maximum allowable hydrogen of 4vol% on 3-second rolling average and 8vol% at any time that was set down in HFCV-GTR Phase 1. Under these conditions, combustion behavior of the air/hydrogen premixed gas and thermal influence around the fuel cell exhaust outlet were investigated.

3.1. Test method

Figure 6 shows the schematic of the exhaust pipe model used in this test. While this model was similar to the model shown in Figure 1, an extra bend was added to the 1-inch pipe while metallic meshes were inserted into the pipes in order to facilitate mixing of sporadically released hydrogen gas into a continuous stream of air inside the exhaust pipe. Horizontal and downward discharge directions were adopted as two test conditions, and the downwardly directed exhaust outlet was positioned 150 mm high from the ground. Air flow rates of 50 NL/min and 500 NL/min, an oxygen concentration of 20.8vol%, and a humidity of 0RH% were applied. Since this test did not set humidity as a parameter, the bubbling chamber was excluded from the exhaust pipe model. A three-way valve was employed to constantly discharge hydrogen gas into the surroundings while sporadically releasing hydrogen gas into the exhaust pipe for a duration of several seconds at intervals. Whether or not ignition had occurred was judged from images taken by an infrared thermal imaging camera according to the test method aforementioned in 2.1.

Figure 7 shows the grid of 25 Gardon heat flux gauges employed to measure heat fluxes in ignition instances so as to evaluate burn injury possibility to persons present near the FCV exhaust outlet. As referential data, flame propagation to quantitative filter paper (no.5A of Advantec make) was examined.

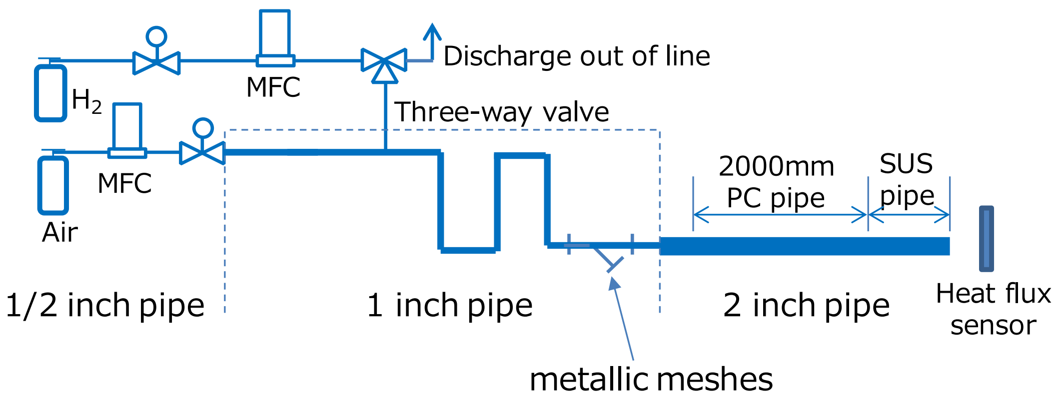

Figure 6. Schematic of exhaust pipe model

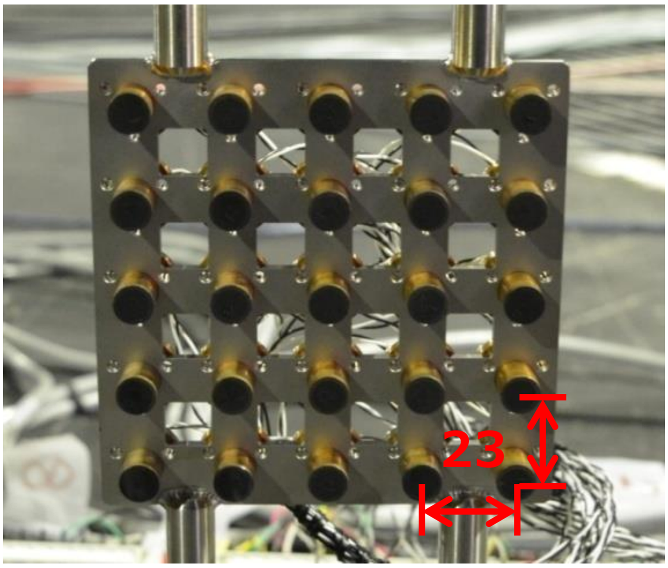

Figure 7. Heat flux sensors

3.2. Test results and discussion

Figures 8 and 9 show infrared thermal imaging camera views of ignition behavior at the exhaust outlet in horizontal and downward directions under the condition where the air flow rate was 500 NL/min, the hydrogen concentration of air/hydrogen premixed gas 3.5vol% on 3-second average and no more than 8.6vol% at any time.

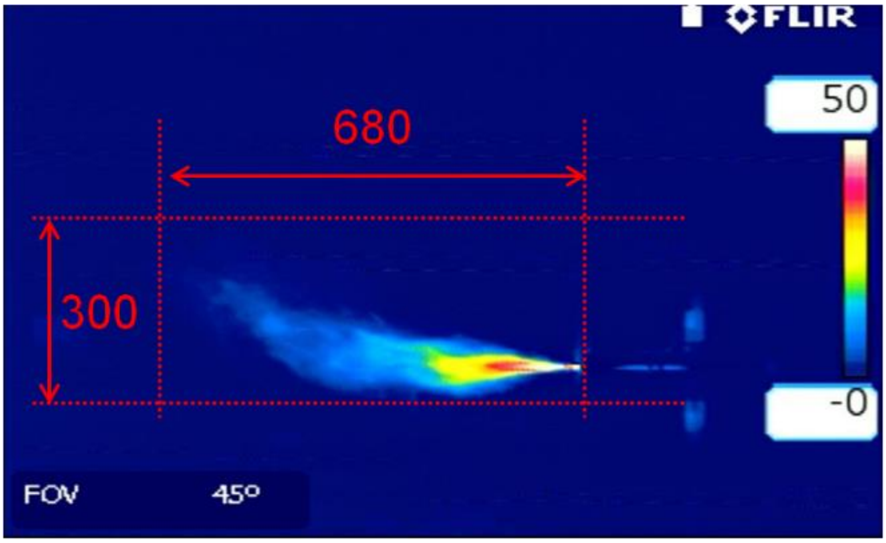

Figure 8. Ignition behavior (side view)
(Air 500NL/min, Horizontal discharge)

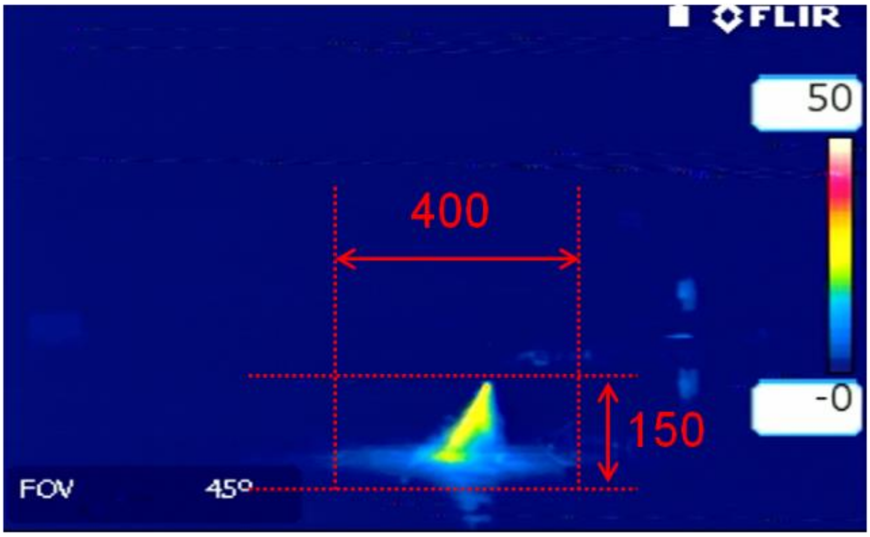

Figure 9. Ignition behavior (side view)
(Air 500NL/min, Downward discharge)

In the case of horizontal discharge, an area 680 mm in length and 300 mm in height revealed temperature rise due to combustion. On the other hand, downward discharge resulted in temperature rise in an area 400 mm in length and 150 mm in height. Horizontal discharge created a greater temperature rise area than did downward discharge whose flame range was limited by the ground.

Figure 10 shows an infrared thermal imaging camera view of ignition behavior at the exhaust outlet in horizontal direction under the condition where the air flow rate was 50 NL/min, the hydrogen concentration of air/hydrogen premixed gas was 6.8vol% on 3-second average and no more than 13.8vol% at any time. These hydrogen concentrations were much higher than those prescribed in HFCV-GRT Phase 1.

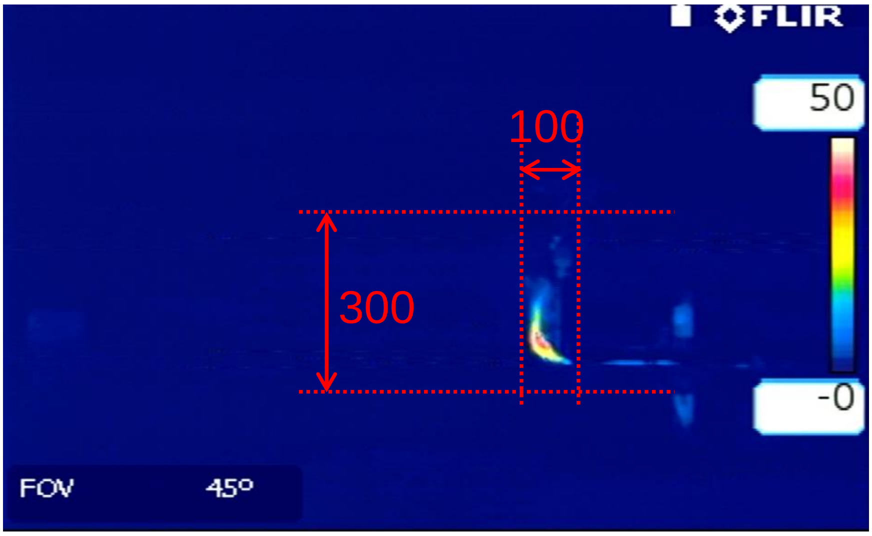

Figure 10. Ignition behavior (side view)
(Air 50NL/min, Horizontal discharge)

The test results indicated that with an air flow rate of 50 NL/min, temperature rose in an area 100 mm long and 300 mm high. Comparison with the results in Figure 8 indicated that a lower flow rate of purged gas generated a smaller area of temperature rise even though the hydrogen concentration in air/hydrogen premixed gas was higher. When the air flow rate was kept at 50 NL/min but the hydrogen concentration was lowered to 4vol% on 3-second rolling average and no more than 8vol% at any time, there were mixed cases of ignition occurring and not occurring.

Next, Figure 11 shows burn standings based on the calculation of heat fluxes measured under Figures 8~9 conditions (i.e., 500 NL/min, 3.5vol% on 3-second average, no more than 8.6vol% at any time, horizontal and downward discharge) and also under Figure 10 conditions (i.e., 50 NL/min, 6.8vol% on 3-second average, no more than 13.8vol% at any time, horizontal discharge). Burn evaluation was carried out using the Equation(1) developed by Eisenberg, N.A. et.al.[5], whereby cases were to be classified as "no first-degree burn" if the integral values of heat flux measurements did not exceed 550,000. "First-degree burn" is defined as a burn accompanied with rash and pain but curable within days, as established by the academic society of Japanese dermatologists[6].

$$tI^{1.15} = 550000 \qquad (1)$$

I = heat flux value (W/m^2), t = time (s)

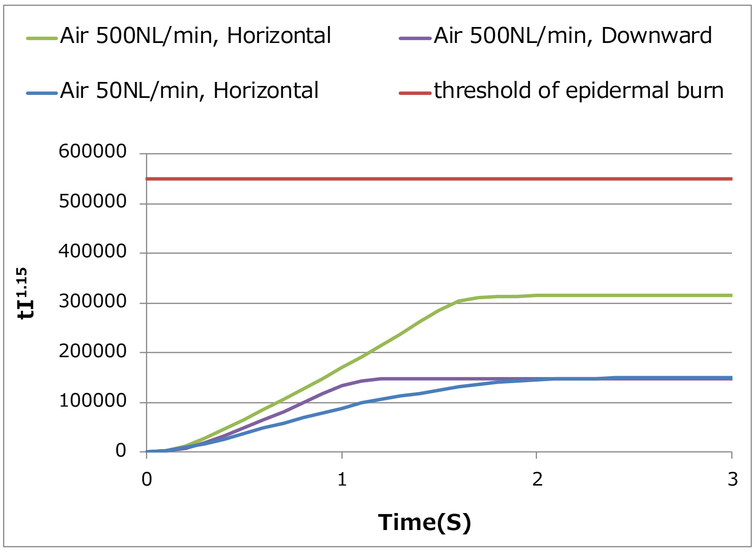

Figure 11. Evaluation of burn

It was found that under the test conditions for Figure 11 none of persons present in the vicinity of the fuel cell exhaust outlet sustained first-degree burn from the combustion of purged gas. Accordingly it was concluded that the hydrogen concentration limit of purged gas prescribed in HFCV-GTR Phase 1 is clearly valid from the standpoint of safety.

4. CONCLUSIONS

To reevaluate the ignition conditions and associated risks of purged gas in relation to the maximum allowable hydrogen concentration prescribed in HFCV-GTR Phase 1, combustion behavior and safety at a time of ignition were investigated under test conditions approximating the actual hydrogen gas purging conditions for FCV. Concerning one of the assumptions applied in HFCV-GTR Phase 1 that there is no flame holding below 8% hydrogen, the present test results indicated that flame holding could occur above 8% hydrogen only when the air flow rate was approximately 2,000 NL/min, and that flame holding could take place below 8% hydrogen when the air flow rate was 50 NL/min or 500 NL/min. Thus, compared with flow rate, oxygen concentration and humidity proved less influential on the ignition behavior of purged gas. In addition, the test results indicated that ignition sometimes failed to occur in air/hydrogen premixed gas at a hydrogen concentration of approximately 4vol% on 3-second average and 8vol% at any time, and that even if ignition occurred at slightly higher hydrogen concentrations, its thermal effect was so limited that no burn was inflicted upon persons nearby while no flame was propagated to quantitative filter paper. It was therefore concluded that hydrogen concentration in air/hydrogen premixed gas specified in HFCV-GTR Phase 1 is valid.

5. ACKNOWLEDGMENTS

This study was carried out as part of the “research and development project for the harmonization of hydrogen safety standards for fuel cell vehicles” commissioned by New Energy and Industrial Technology Development Organization (NEDO), a national research and development agency of Japan.

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