DISPERSION TESTS ON CONCENTRATION AND ITS FLUCTUATIONS FOR 40MPa PRESSURIZED HYDROGEN

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ABSTRACT

Hydrogen is one of the important alternative fuels for future transportation. At the present stage, research into hydrogen safety and designing risk mitigation measures are significant task. For compact storage of hydrogen in fuel cell vehicles, storage of hydrogen under high pressure, up to 40 MPa, at refueling stations is planned, and safety in handling such high-pressure hydrogen is essential. This paper describes our experimental investigation into dispersion of high-pressure hydrogen gas which leaks through pinholes in the piping to the atmosphere.

First, in order to comprehend the basic behavior of the steady dispersion of high-pressure hydrogen gas from the pinholes, the time-averaged concentrations were measured. In our experiments, initial release pressures of hydrogen gas were set at 20 MPa or 40 MPa, and release diameters were in the range from 0.25 mm to 2 mm. The experimental results show that the hydrogen concentration along the axis of the dispersion plume can be expressed as a simple formula which is a function of the downwind distance X and the equivalent release diameter θ. This formula enables us to easily estimate the axial concentration (maximum concentration) at each downstream distance.

However, in order for the safety of flammable gas dispersion to be analyzed, comparisons between time-averaged concentrations evaluated as above and lower flammable limit are insufficient. This is because even if time-averaged concentration is lower than the flammability limit, instantaneous concentrations fluctuate, and a higher instantaneous concentration occasionally appears due to turbulence. Therefore, the time-averaged concentration value which can be used as a threshold for assessing safety must be determined considering concentration fluctuations. Once the threshold value is determined, the safe distance from the leakage point can be evaluated by the above-mentioned simple formula. To clarify the phenomenon of concentration fluctuations, instantaneous concentrations were measured with the fast-response flame ionization detector. A small amount of methane gas was mixed into the hydrogen as a tracer gas for this measurement. The relationship between the time-mean concentration and the occurrence probability of flammable concentration was analyzed. Under the same conditions, spark-ignition experiments were also conducted, and the relationship between the occurrence probability of flammable concentration and actual ignition probabilities were also investigated. The experimental results show that there is a clear correlation between the time-mean concentration, the occurrence probability of flammable concentration, flame length and occurrence probability of hydrogen flame.

NOMENCLATURE

a_1: Proportional constant
C: Instantaneous concentration
C_m: Time-averaged concentration

D: Nozzle diameter of a leakage opening
L_F: Flame size
L_D: Flame propagation distance from ignition point: L_D
P_0: Hydrogen initial pressure
P_C: Occurrence probability of flammable concentration
P_F: Occurrence probability of hydrogen flame
PDF: Probability density function
X: Distance from the leakage nozzle
Z: Height above the ground
θ: equivalent release diameter
ρ_0 is hydrogen density at nozzle throat
ρ_a is density of ambient air.

1.0 INTRODUCTION

Hydrogen refueling stations which are being planned at the present will store high-pressure hydrogen gas at 40 MPa, and the acquisition of basic data on the influence of high-pressure hydrogen gas on the surroundings has become an urgent task. In this research we focused on a typical leakage scenario, i.e., leakage from a pinhole occurring in equipment, resulting in continuous leakage at a constant mass flow rate (steady leakage).

Our research has two main objectives. One is to comprehend the basic behavior of the dispersion of high-pressure hydrogen gas where there is steady leakage from a pinhole. Field dispersion experiments were conducted for this purpose, and data sets of time-averaged concentrations were obtained and analyzed.

The other objective is to study the flammability of the time-dependant concentration fluctuations. To investigate the safety of flammable gas dispersion, time-averaged concentrations are insufficient. This is because even if time-averaged concentration is lower than the flammability limit, instantaneous concentrations fluctuate and a higher concentration occasionally appears due to turbulence [1]. Considering this, threshold concentration values for determining safety are often set to lower than LFL (lower flammable limit), for example 1/2LFL in U.S.A.[2] or 1/4LFL in Japan[3]. However, there has not been much research on concentration fluctuation and flame propagation in the atmosphere under high-pressure leakage conditions. To accomplish this objective of our research we must understand the relationships among the concentration fluctuation, the occurrence probability of flammable concentration and the features of the flame propagation. To clarify these relationships, further experiments were conducted. In these experiments, instantaneous concentration fluctuations of hydrogen from pinhole leakage were measured. Spark-ignitions were also applied at the same point simultaneously with such fluctuations, and the characteristics of the flame propagation were analyzed.

2.0 DISPERSION EXPERIMENTS FOR TIME-AVERAGED CONCENTRATIONS

2.1 Experimental Apparatus and Test Field

Field dispersion experiments of high-pressure hydrogen were conducted at the Tashiro testing facility of Mitsubishi Heavy Industries, Ltd. in order to ascertain the characteristics of time-averaged concentrations where there is pinhole leakage.

Hydrogen gas was first stored in five high pressure cylinders, each with a capacity of 50 liters, using a pressure booster. The cylinders were connected to a release nozzle (i.e. pinhole) by means of 25 mm diameter piping and a pressure regulator. A valve for control of gas release was equipped at the end of the piping immediately ahead of the nozzle. Considering the pressure loss between the high-pressure cylinders and the nozzle, the pressure of the hydrogen gas in the cylinders was set at 65 MPa. The diameters of the release nozzles prepared were D= 0.25 mm, 0.5 mm, 1.0 mm and 2.0 mm and the release pressure, P_0, were 40 MPa and 20 MPa. The test conditions were as follows.

(1) P_0= 40 MPa: D= 0.25 mm, 0.5 mm, 1.0 mm and 2.0 mm
(2) P_0= 20 MPa: D= 2.0 mm

In each test, the release point was 1.0m above the ground level and the release direction was horizontal. The thermal conductivity type gas sensors KD-3A, produced by New Cosmos Electric Company, were used for measuring time-averaged hydrogen gas concentrations. The sensors were mounted on poles located at X=4, 6, 8, 11, 15, 20, 30 and 50m, where X is the distance from the leakage nozzle. 5 sensors were mounted on each pole for the purpose of measuring the vertical concentration profile. The coordinates of each sensor are shown in Figure 1. The meteorological conditions, wind speed and wind direction, were also measured, with an ultrasonic anemometer at the location X=-50m.

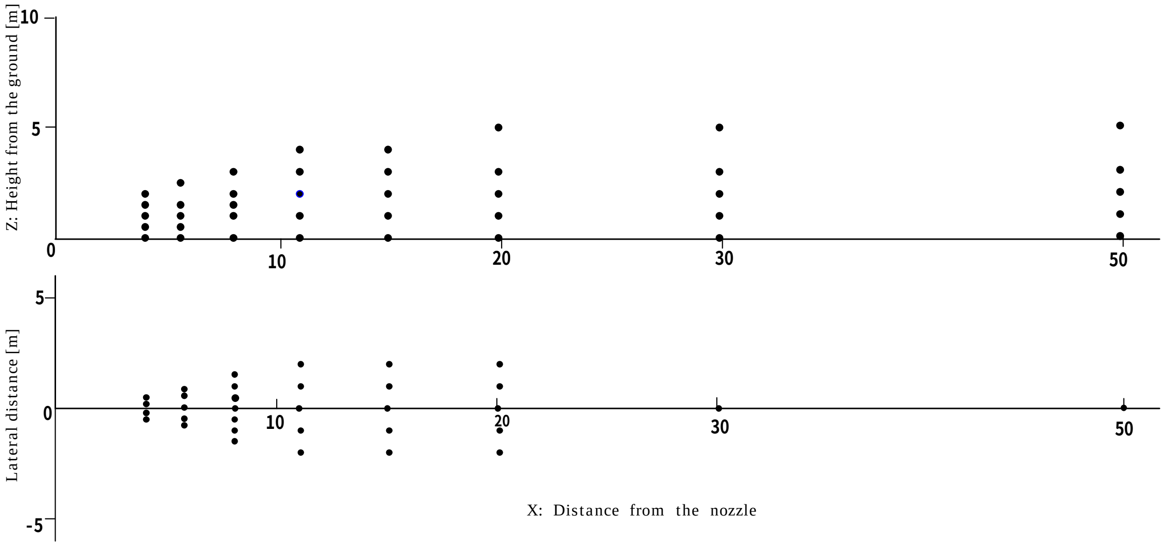


Figure 1. Coordinates of hydrogen gas concentration sensors

2.2 Results and Discussion

Figure 2 shows a typical example of concentration contour under the leakage conditions P_0=40MPa and D=2.0 mm. As can be seen, the gas plume is almost horizontal near the leakage point, and showing that the momentum effect is more dominant than the buoyancy effect of hydrogen in a high concentration area. Therefore, using an analogy from the turbulent jet characteristics of incompressible jet flow, we plotted the time-averaged concentration against X/θ along the axis of the dispersion plume. The parameter θ is the equivalent release diameter defined as below,

$$\theta = D\sqrt{\frac{\rho_0}{\rho_a}} \tag{1}$$

where ρ_0 is hydrogen density at nozzle throat, ρ_a is density of ambient air. The result is shown in Figure3. At larger X/θ, i.e., far from the release point, scattering of concentration is relatively larger due to the fluctuation of meteorological conditions and smaller momentum of hydrogen jet. However, at a short distance where the effect of fluctuation of atmospheric conditions is relatively small, the scattering becomes smaller and the plotted points are almost in alignment with a line indicating an inverse linear relation between C_m and X/θ. In this way, we can see that the time-averaged hydrogen concentration along the axis of the dispersion plume can be expressed as a simple formula as shown below, where the value of proportional constant a_1 is estimated to be 6000[4]. This formula will enables us to estimate the axial concentration at any distance easily in case the release diameter and initial pressure are given.

$$C_m = a_1 \left(\frac{X}{\theta} \right)^{-1} \tag{2}$$

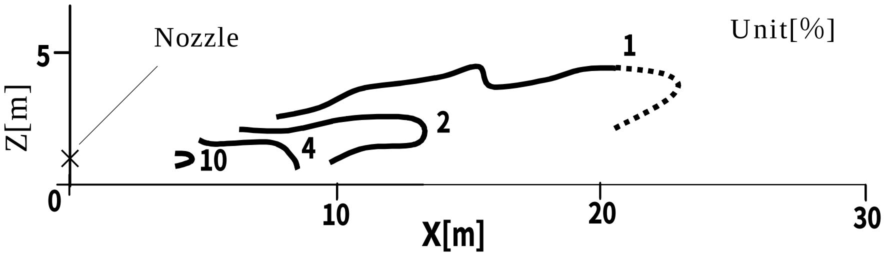


Figure 2. Time-averaged concentration contour (P_0=40MPa, D=2.0mm)

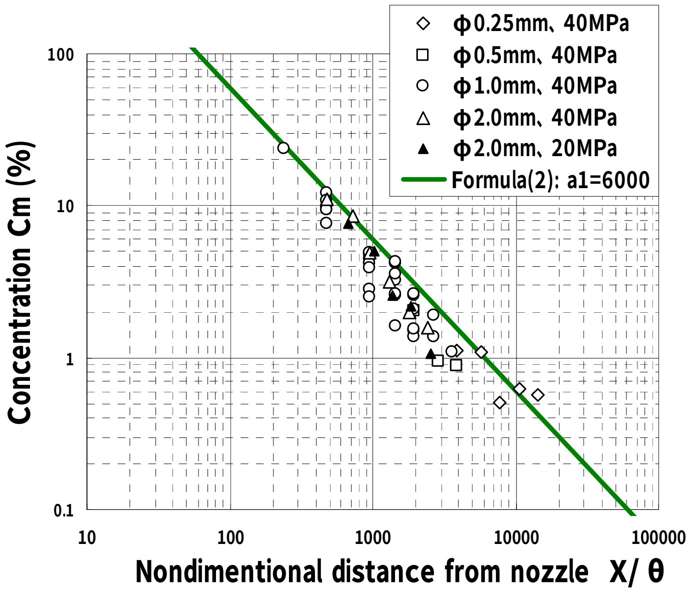


Figure 3. Concentration distribution

3.0 DISPERSION AND SPARK-IGNITION EXPERIMENTS FOR CONCENTRATION FLUCTUATIONS

3.1 Apparatus and the Method of the Experiments

Field dispersion and ignition experiments of high-pressure hydrogen were conducted at the test facility in the Nagasaki Research & Development Center of Mitsubishi Heavy Industries, Ltd. In these experiments, we focused on the concentration fluctuation and the characteristics of flame propagation. A schematic diagram of the experiments is shown in Figure 4. Hydrogen pressure was boosted up to 40MPa and released horizontally as pinhole leakage. The diameter of the pinhole was 0.2mm. Methane gas was mixed into hydrogen gas to a concentration of 1.5vol%, for the purpose of measuring concentration fluctuation using a fast response flame ionization detector (FID, HFR-400 by CAMBUSTION). Since response speed of conventional hydrogen gas sensors is usually low (several seconds), they are inappropriate for detecting the concentration fluctuations (i.e. instantaneous concentration). On the other hand, using fast response FID, a high frequency of measurements of methane gas concentration could be achieved. Therefore, instead of measuring hydrogen gas concentration itself, concentration fluctuations of methane gas were measured firstly and then they were converted into concentration of hydrogen using the factor of 100/1.5. The sampling rate of the FID was 200 Hz.

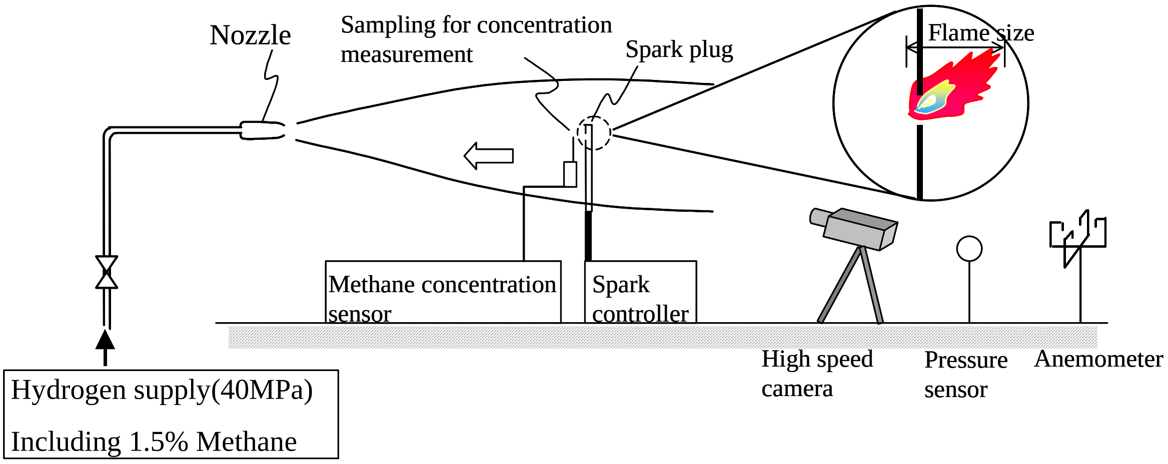


Figure4. Schematic diagram of the experiment

In these experiments, electric spark-ignition was also applied simultaneously with concentration measurements at the vicinity of the point of concentration measurement. Frequency of the spark was controlled to 10 Hz, which was slow enough for instantaneous flame generated around the spark to go downstream and disappear before the next spark. The electrode gap size is 1.5 mm and the electric energy of each spark was set to 120 mJ, which is much larger than the minimum ignition energy of hydrogen.

The phenomena of flame generation and propagation were recorded with a 500f/s high speed video camera (FASTCAM-APX, Photron) and analyzed by image processing. An UV band-pass filter (313 ± 7 nm) was attached to the high speed video camera, making it possible to take images of the hydrogen flame, emission with a spectrum whose peak was around 310 nm. Ambient pressure change due to ignition was also measured with a pressure sensor (XCS-062: KULITE SEMICONDUCTOR) located 2.5m away from the nozzle. Wind speed and wind direction were measured with an ultrasonic anemometer. Measurement duration per point was about 40-50 seconds. During this time the data for the time history of instantaneous concentration was taken at a rate of 200 Hz, and about 400 ignition images were obtained. The experimental conditions are summarized in Table1.

Table 1. Summary of experimental conditions.

Release condition	Release pressure: P_0	40MPa
	Nozzle diameter: D	0.2mm
Measurement point	Distance from the nozzle: X	0.15, 0.3, 0.35, 0.4, 0.45, 0.7, 0.8, 0.9, 1.0, 1.5, 2.0m

Obtained data was analyzed as shown below.

(1) Concentration time-history data analysis: The time history of instantaneous concentrations at each measurement point was analyzed statistically to obtain the following parameters.
a) Time-averaged concentration: C_m
b) Probability density function of concentration: *PDF*
c) Occurrence probability of flammable concentration: P_C (i.e. the probability of occurrence of concentrations ranging from 4% to 75%)

(2) Image data analysis: For each image from the high speed video camera, the size of hydrogen flame was analyzed to obtain the following parameters at each measurement point.
a) Occurrence probability of hydrogen flame: P_F (= number of times flame was generated/ number of times sparks were generated)
b) Flame size : L_F (Maximum size during one spark)
c) Flame propagation distance from ignition point: L_D (See Figure 5)

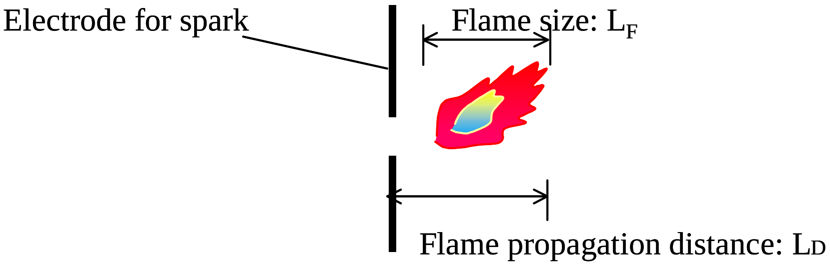


Figure5. Definition of L_F and L_D

3.2 Results and Discussion

(1) Results of concentration measurement

Figure 6 shows a typical example of concentration measurement at X=0.9m, where time-averaged concentration is 4.1%. As can be seen, instantaneous concentration fluctuates and is higher than the time-averaged concentration sometimes appears. The probability density function of concentration derived from this time-history is shown in Figure 7. The probability distribution has a peak close to the time-averaged concentration but concentration higher than 6% also appears. Figure 8 is the distribution of time-mean concentration with the variation in distance from the nozzle.

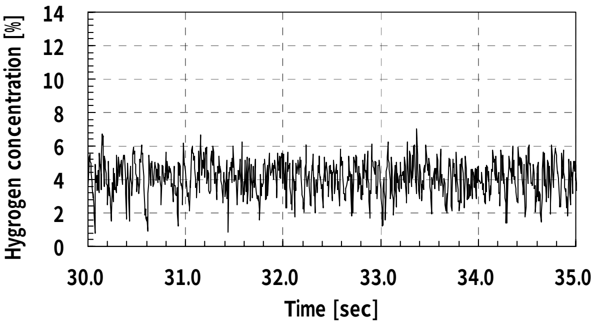


Figure6. Concentration time history (X=0.9m)

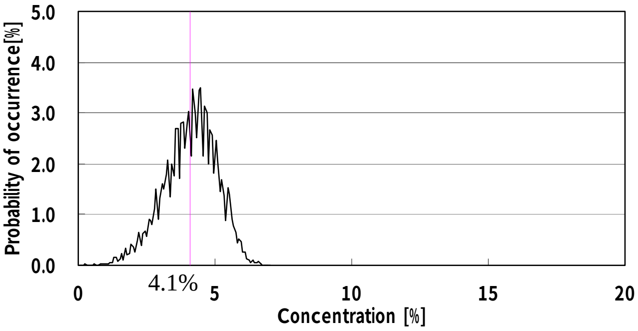


Figure7. Probability density function (X=0.9m)

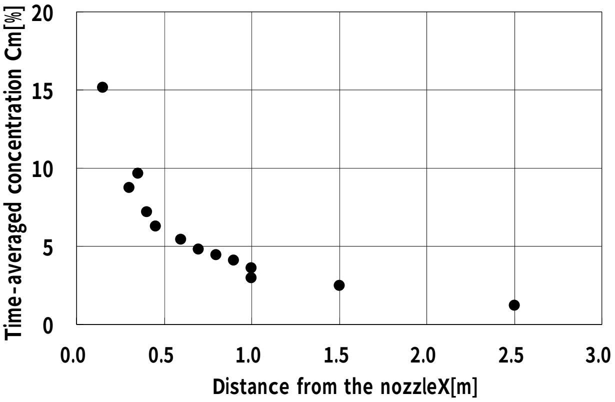


Figure8 Distribution of time-mean concentration

The relationship between time-averaged concentration C_m and the occurrence probability of flammable concentration P_C is shown in Figure9. If C_m is larger than about 7%, Pc has a value near 100%. This means the concentration is always higher than lower flammable limit, i.e. 4%. On the other hand, as C_m becomes smaller, *Pc* decreases and it becomes almost zero at around C_m =2%. This implies that there is no possibility of ignition in an area where time-averaged concentration is lower than 2%.

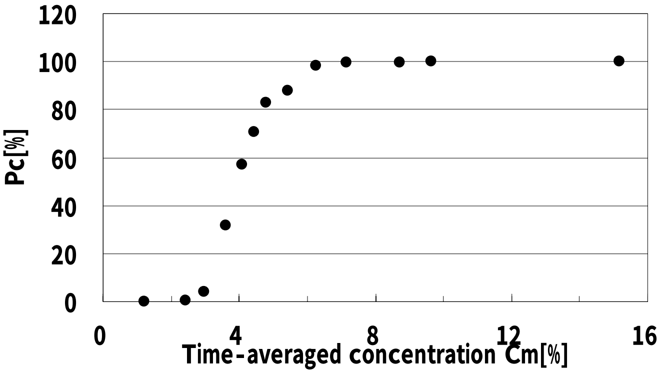
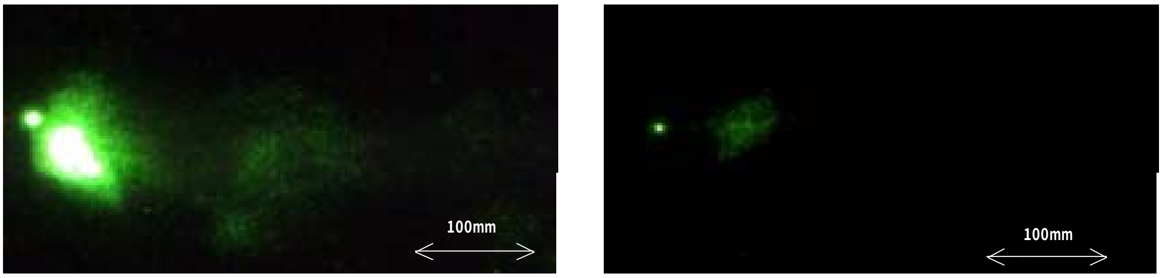


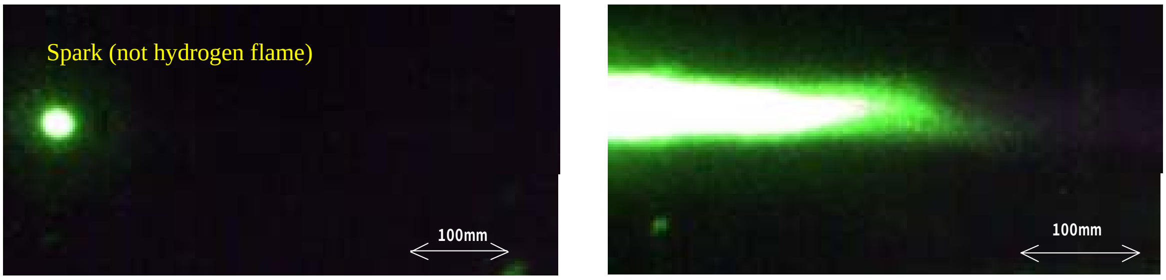
Figure 9 Relationship between C_m and the P_C

(2) Results of spark-ignition experiments

Figure 10 shows a typical example of images from the high-speed camera of ignition experiments at X=0.15m, 0.3m, 0.6m and 0.9m. At X=0.3m, where time-averaged concentration is 8.7% almost twice the lower flammable limit, the size of hydrogen flame in this image is about 300 mm, and at X=0.6m, where time-averaged concentration is 5.4%, the size becomes smaller. On the other hand, at X=0.9m, where time-averaged concentration is 4.1%, hydrogen flame cannot be recognized. Here it seems that a flame was not generated or if it was, the flame size was smaller than the spark size i.e., smaller than a few millimeters. On the contrary, at the point of much higher concentration, X=0.15m, where time-averaged concentration is 15.2%, the flame was generated at the first spark and it grew to a steady jet flame which had a length of 450 mm. The relationship between time-averaged concentration and flame size is shown in Figure 11. In this graph, the square symbols represent the maximum L_f value among all the sparks at each measuring point, and circle symbols represent the averaged values. Figure 12 also shows the relationship between occurrence probability of hydrogen flame (P_F) and time-averaged concentration, where the P_F is defined as a ratio of the number of times flame larger than the spark size was generated to the number of times sparks were applied. As can be seen, in case time-averaged concentration is large enough, around 15%, flame propagated upstream to the nozzle and it formed a steady state jet flame. On the other hand as time-averaged concentration decreases, both flame size and P_F become smaller, approaching zero around at C_m=5%. This phenomenon is consistent with the fact that Pc is almost zero when C_m is around 2%, i.e., when there is no possibility of ignition. In every measurement during the experiments, ambient pressure change caused by flame generation was at an undetectable level, i.e. less than 100 Pa. These facts imply that under these release conditions and in the area where time-averaged concentration is lower than 2% at least, no significant flame propagation occurs. However, these results were derived from experimental data under regulated conditions. Further data accumulation under various conditions such as larger nozzle diameter or where there are obstacles is to be expected and this will be our future task.



(a)X=0.3m (C_m=8.7%, P_C=99.8%)   (b)X=0.6m (C_m=5.4%, P_C=88%)



(c)X=0.9m (C_m=4.1%, P_C=57%)   (d)X=0.15m (C_m=15.2%, P_C=100%)

Figure10 Examples of images from high-speed camera

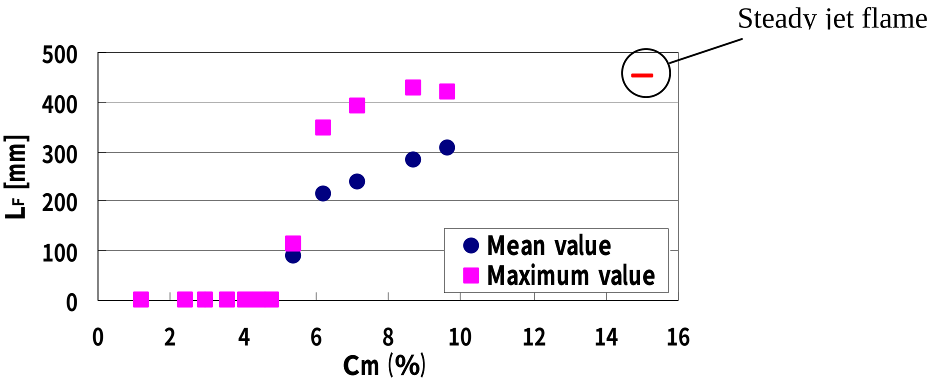


Figure11. Relation ship between the time-averaged concentration and flame size

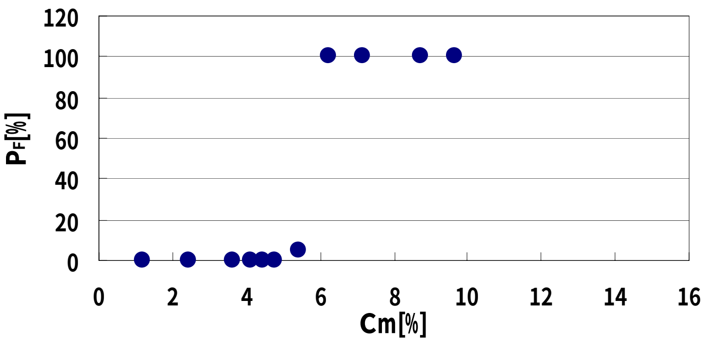


Figure12. Relation ship between the time-averaged concentration and P_F

4.0 CONCLUSIONS

To comprehend the basic behavior of the dispersion of high-pressure hydrogen by pinhole leakage, field dispersion experiments were conducted. The initial release pressure of hydrogen gas were set at 20 MPa or 40 MPa, and release diameters were in the range from 0.25 mm to 2 mm. Data sets of time-averaged concentrations were obtained and analyzed. The experimental results show that time-averaged hydrogen concentration along the axis of the dispersion plume can be expressed by a simple formula as shown below, where the value of this proportional constant a_1 is estimated to be 6000. This formula will enable us to estimate axial concentration at each distance easily.

$$C_m = a_1\left(\frac{X}{\theta}\right)^{-1} \quad (2)$$

To evaluate the safety of dispersed flammable gas, however, comparisons between time-averaged concentrations and lower flammable limit are insufficient because of concentration fluctuations. Dispersion and spark-ignition experiments of high-pressure hydrogen were conducted for the purpose of clarifying the relation between concentration fluctuations and the flame propagation. The experimental results show that the occurrence probability of flammable concentration decreases with decrease in the time-averaged concentration and becomes almost zero and no significant flame propagation occurs, where C_m is around 2% or less. Thus, the experimental results show that there is a clear correlation between the time-mean concentration, the occurrence probability of flammable concentration, flame length and occurrence probability of hydrogen flame. However, these results were derived from experimental data under regulated conditions. Further data acquisition under various conditions such as larger leakage diameter or where there are obstacles is to be expected and this will be our future task.

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