Submission ID:230

3D real time monitoring of H2 in FCV applications

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Abstract:

In order to monitor a trace amount of Hydrogen in millisecond, portable H_2 sensor (Sx) was made by using mass spectrometer. The method of monitoring the hydrogen pulse of millisecond in exhaust gas is the increasing needed. Determining H_2 concentration both inside and outside of the Fuel Cell Vehicle (FCV) for the optimized operations is becoming a critical issue. The exhaust gas of Fuel Cell Vehicle, H_2 consumption, flushing and disposal around Fuel cell, the real time monitoring of H_2 in highly humid conditions is the problematic. To solve this issue, the system volume of the sampling route was minimized with the heater and the dehumidifier to avoid condensation of water droplets. And also for an automatic calibration of H_2 concentration, the small cylinder of specific H_2 concentration was mounted into the system.

Our basic experiment started from a flow pattern analysis by monitoring H_2 concentration in narrow tube. The flow patter analysis was carried out. When H_2 gas was introduced in the N_2 flow or air in the tube, the highly concentrated H_2 front phases were observed. This H_2 sensor can provide the real time information of the hydrogen molecules and the clouds. The basic characterization of this sensor showed 0-100% H_2 concentrations within milliseconds. Our observations showed the size of the high concentration phase of H_2 and the low concentration phase after mixing process. The mixed and unmixed H_2, unintended concentration of H_2 cloud, the high speed small cluster of H_2 molecules in purged gas were explored by this system.

1. Introduction

Towards 2020 Tokyo Olympic Game, various Hydrogen applications without CO2 emission are prospectively being developed. Fuel-cell in house application is expected to simultaneously generate tri-generation of electricity, heat and hot water. Fuel Cell Vehicles (FCV) in new efficient models with the dramatically optimized in mechanical, electrical, and chemical background technologies come out. From the safety point of view, the explosion limit for H_2, 4% and GTR(Global technical regulation) on FCV considering the value of 4% and 8% of H2 release. In production line, the H_2 concentration limit of 0.1% and 1% are the comprehensible upper limit. By GTR, Fuel cell discharge system at the vehicle exhaust system`s point of discharge, the hydrogen concentration level shall not exceed 4 % average. Our focus is to analyze the mixing process of 100% H_2 with air to find unintended concentration of H_2 in clouds. The real time monitoring of the process of mixing 100% H_2 with air in milliseconds is our study purpose. Considering real time monitoring of H_2 concentration, The portable hydrogen sensor (Sx) is built using mass spectrometer to measure gas mixtures in millisecond intervals. Several Sxs are located to measure the real time images of 3-Demensional H_2 concentrations. The concentration profile in milliseconds by high-speed camera is taken using the Schieren technique at the same time.

GTR for H_2 disposal; Agreement concerning the establishing of GTR for wheeled vehicles, equipment and parts which can be fitted and/or be used on wheeled vehicles is mostly solidified. According to GTR on FCV, fuel cell discharge system at the vehicle exhaust system`s point of discharge, the hydrogen concentration level shall not exceed 4 % average by volume during any moving three-second time interval during normal operation including start-up and shut down[1]. FC stack needs to be washed out by the concentrated hydrogen as the purge gas and how to

exhaust gas without exceeding 4 % is the most concerns [9]. Also how to measure hydrogen pulse of millisecond in exhaust is the rising up issue. Further more, any single failure downstream of the main H_2 shut off valve shall not result in a H_2 concentration in air of 4 % or more by volume within the passenger compartment. If a single failure down stream of the main hydrogen shut off valve results in a H_2 concentration in air of 4 % by volume within an enclosed or semi-enclosed space in vehicle, the main H_2 shutoff valve shall be closed and warning to the driver shall be provided. [8] In this paper, model of FCV hydrogen discharge system was composed of plastic tube with pressure gage, Mass Flow controllers and Highspeed Solenoid valves. Variety of simple experiments, injection, mixing, change flow rate and change tube inside diameter were carried out to control the H_2 concentration also Nitrogen (N_2) instead of Air. In mixing experiment, H_2 gas was introduced in the N_2 & Air flow to form the various H_2 concentrations. H_2 at the point of discharge was monitored by the real time H_2 monitoring system Sx. The fast solenoid valve was opened within milliseconds to add H_2 gas in N_2 & Air flow, H_2 gas ran through the tube by mixing with N_2, and the wave front at the point of discharge was observed. In the milliseconds following the mixing process the separated phases were shown by the real time H_2 sensor and the wave front concentration of H_2 during stop and release type of motions was topics. In milliseconds diffusion mechanism does not work but Reyleigh-Taylor instability might work [10].

In order to detect low concentration of H_2 in real time at the same time other gases, various kinds of hydrogen detectors exist but each sensor presents difficulty to measure 0-100 % concentration of hydrogen. Semiconductor gas sensor, for example, cannot measure 100 % hydrogen. Catalytic gas sensor with filter needs time for hydrogen to penetrate filter. Neither laser detector does not work in millisecond and humid condition. In order to detect hydrogen in real time, mass spectrometer system with differential pumping stage was selected to develop real time monitoring system and applied to H_2/N_2 mixing experiments [5,6]. In hydrogen release experiment on the mountain, high pressure H_2 gas was emitted from the pipe in the air to form the various H_2 concentrations of cloud. H_2 diffusion process was monitored by the real time H_2 monitoring system [5, 7,11,12].

2. Experimental

2-1 Sx H_2 Sensor

Photo images of Sx H_2 Sensor and outdoor experiment on the mountain are shown in Fig. 1&2.

H_2 Sensor (Sx) is 25kg and portable for the outdoor experiment. Sx has self-calibration system having certain concentration of single and mixed calibration gases for easy start up. Sx can take data each 2ms intervals. Sampling inlet of this system is SUS capillary tube (150/250μm=ID/OD). Sample gas is introduced from capillary through dehumidifier. This system having differential pumping system, ionize by electron bombardment method (EB). To minimize the humidity influence, Sx has dehumidifier with heater. Mass Spectrometer works up to m/Z=300. Schlieren Photograph (SP) is applied to monitor for mixed gases density profile. So from gas exit we can monitor the concentration profile by Sx and density profile by SP at the same time. SP is monitored by high-speed camera in 1 msec.

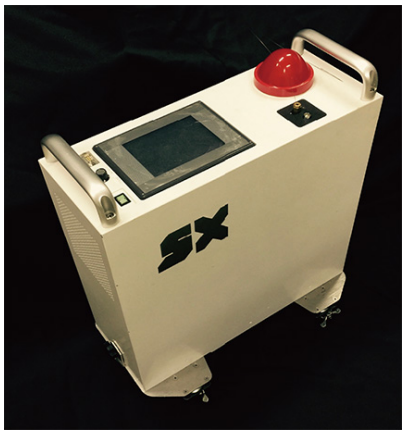

Fig.1 H_2 Sensor(Sx)

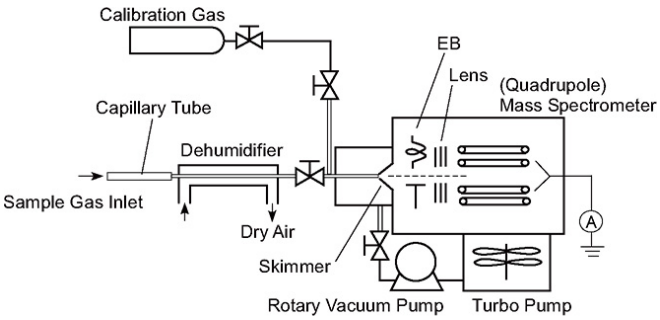

Fig.2 Schematic diagram of Sx

Schematic diagram of Sx is shown in Fig.2. Weight of Sx is 25kg and portable for the outdoor mountain experiment [5]. When Sx measure H_2, alarm red light is on. Liquid crystal touch panel is used to control Sx to start by changing valves. Sx has self-calibration system having certain concentration of single to mixed calibration gases. And having these calibration lines for each gas, Sx can monitor and show the concentration of each gas immediately. Real time monitoring is carried out from 2ms to 30ms. For the real time monitoring system inside volume of Sx is minimized. Sampling inlet of this system is 150-250 μm SUS capillary tube. Gas sample is introduced from capillary through dehumidifier and skimmer to ionization chamber. This system having differential pumping system to analyse gas, ionize hydrogen by electron bombardment method (EB). To minimize the humidity influence, Sx having dehumidifier is heated. Mass Spectrometer system works up to m/Z=300. [7]

2-2 Mixing Hydrogen with Nitrogen or Air by Vertical mixing (right-angle mixing head)

Nitrogen flow is controlled by Mass flow controller (Kofloc Kyoto Model 38100SII-V-1,N_2,In 0.15/out0Mpa,100SLM,20℃,1 atm) with solenoid valve (CKD AB41-02-7 0.25Mpa) to maintain constant flow rate of 5L/min. Hydrogen is added vertically to nitrogen or air flow. At the constant flow rate through Mass flow controller (Kofloc Kyoto Model 3810DS-V, H_2, In 0.20/out0.1Mpa,100SLM,20℃,1 atm) and solenoid valve (CKD AB41-02-7 0.25Mpa) to maintain and check flow rate. H_2 flow start by quick solenoid valve (KOGANEI K2-100SA-09, 0.2-0.5Mpa) in Fig.3 is controlled by pulse generator. 1-100ms intervals pulse start flow by valves into constant air flow and mixing process to be observed.

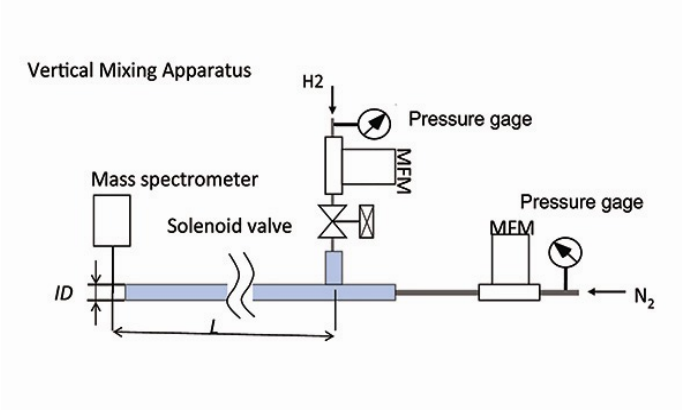

Figure 3. Vertical mixing (right-angle mixing head) equipment with Sx

2-3 H_2/Air Mixing and Combustion/Explosion process by Sx

Experiments are conducted using air instead of nitrogen with ignition equipment. Two tungsten wires are set the head to tale distance of 1cm for the spark where the high voltage of AC12KV is applied. So if H_2 concentration is more than 4%, the ignition process is activated, monitored by SP (Schlieren Photograph) with high-speed camera at the same time 6 Sxs measure the 3D H_2 concentration changes. Explosion data are measured and analyzed.

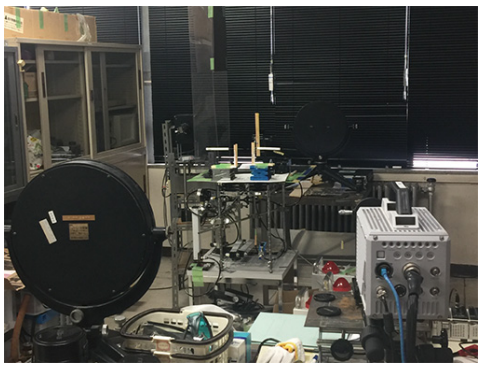

Figure 4. Experimental setup for the mixing and combustion experiment with high speed camera for Schleiren Photograph

Figure 5. Experimental setup of Sxs

2-4 Schleiren Photograph and Sxs

Schleiren Photograph and Sx set up is schematically described below in Fig.6. 3 Sampling capillary position of Sxs has the different height on the exit of the pipe, and each height different by 1cm. The distant between the top of the pipe to the sampling point start from 0.0cm up to 20cm. The photos of Schleiren images were taken by high speed camera.

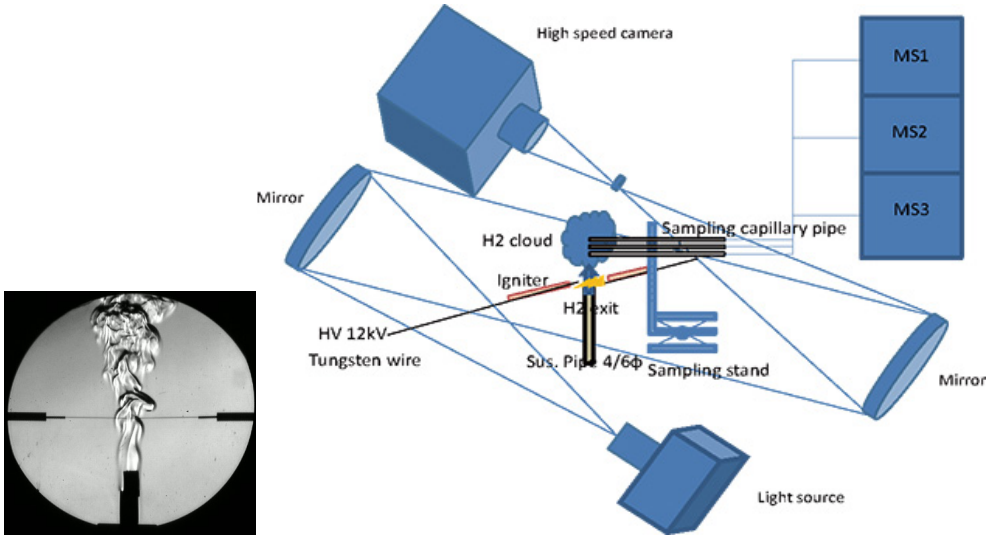

Figure 6. Schematic Diagram of Shleiren Photograph setup

3 Results and Discussions

3-1 Real Time Monitoring of Purged H_2 into Nitrogen flow for 1ms

Under constant flow of N_2 5L/min controlled by massflow controller, pure hydrogen were introduced by quick valve by the interval of 1ms by using vertical mixing with right-angle mixing head shown in Fig.3. The result of H_2 concentration by time was shown in Fig.7. In this experiment single channel was used to monitor H_2 concentration. In this experiment, first hydrogen wave came with the spike head where the highest concentration of hydrogen (Cmax) reached as a cloud by open up quick valve. Hydrogen was introduced into N_2 flow without diffusion and kept the concentration as H_2 cloud. The concentration of spike was more than the expected concentration and above the pre-set value of H_2 4%. Three Sxs (I,O,U) monitored H_2 at the different position of 1,2,3cm from the exit of the pipe, and the maximum concentration H_2 observed near the exit showed 13%. This is more than pre-set value of 4%.

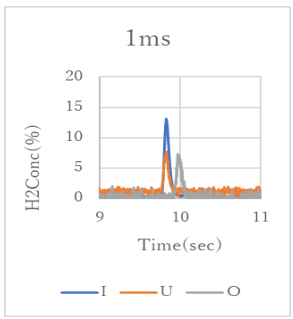

Fig.7 H_2 Response by Quick Valve

3-2 Real Time Monitoring of Purged H_2 into N_2 for 1-100ms

By changing the duration time of quick valve open from 1 to 100ms, the H_2 profile change the shapes and the result are shown in Fig.8. The comparison of H_2 profiles were taken at the same height and showed in Fig.9. At duration time of 1ms, the quick valve motion did not complete open and less volume of hydrogen moved into N_2 flow. The height of spike head reached the maximum concentration of H_2 at the duration time of 10 and keep constant concentrations up to 100ms. When the duration time reach 100ms, the height of spike head became a bit lowere but the width of spike became wider shown in Fig. 8&9. This means that the open to close time of 100ms, hydrogen volume increased compared to lower duration time data shown in Fig. 8&9. At the start point to the maximum concentration of H_2, the slope of the increasing curve was steep compared to the decreasing concentration plofile. This means that the head front of H_2 cloud keep the higher concentration compared to the tail portion. Also this means that more mixing process took place including diffusion in all condition.

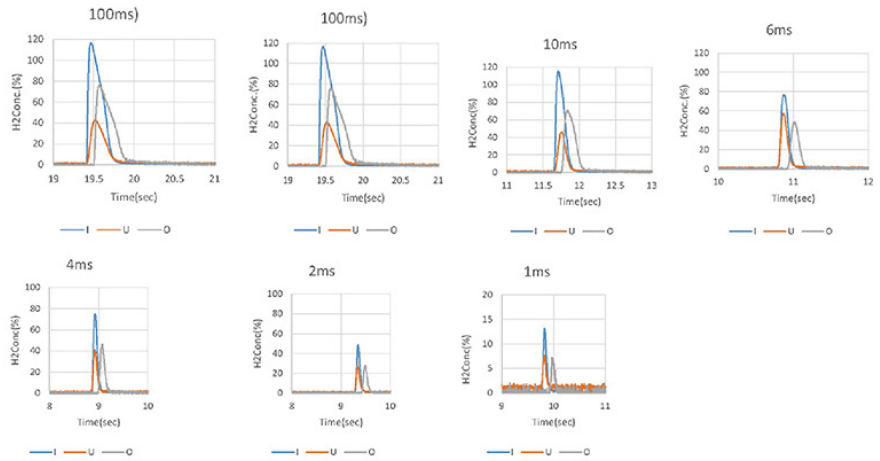

Fig.8 Spike heads changes by response time 1-100m

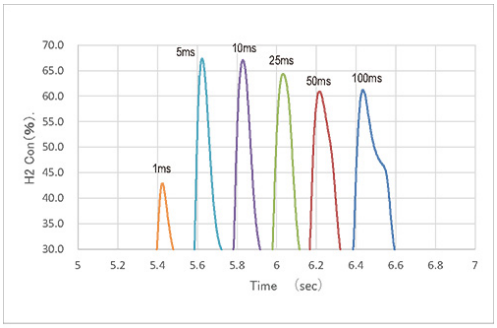

Fig.9 Shape of Spike heads by changing release time 1-100ms.(H_2 Concentration Profile of head3cm of the flow)

By changing the sampling position in 0cm to 20cm for x-axis and 1cm to 3cm in y-axis, hydrogen concentration were monitored through the vertical mixing equipment. All data were plotted and shown in Fig.10. The slope of line shown in this figure explained that the maximum concentration of ejected H_2 clouds decreased with the distance from the exit of pipe and decreased inverse proportionally to the distance between the exit and sampling point. The maximum concentration and the distance L showed the upper slope line in Fig.10.

3-3 Real Time Monitoring of the spike head of H_2 purged by changing position X-Y

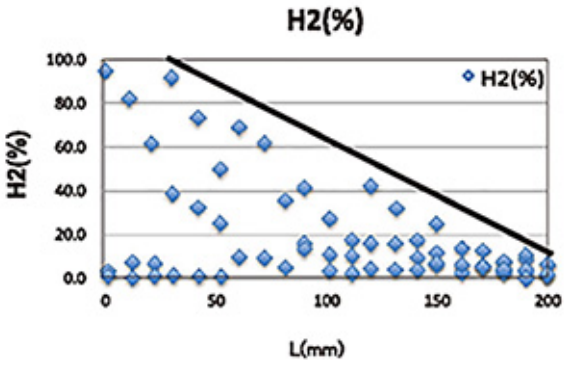

Fig.10 H_2 concentration at Spike head by changing X-Y MS gas sampling positions

; preset H_2 of 4% , H_2 100%100ms release in Air

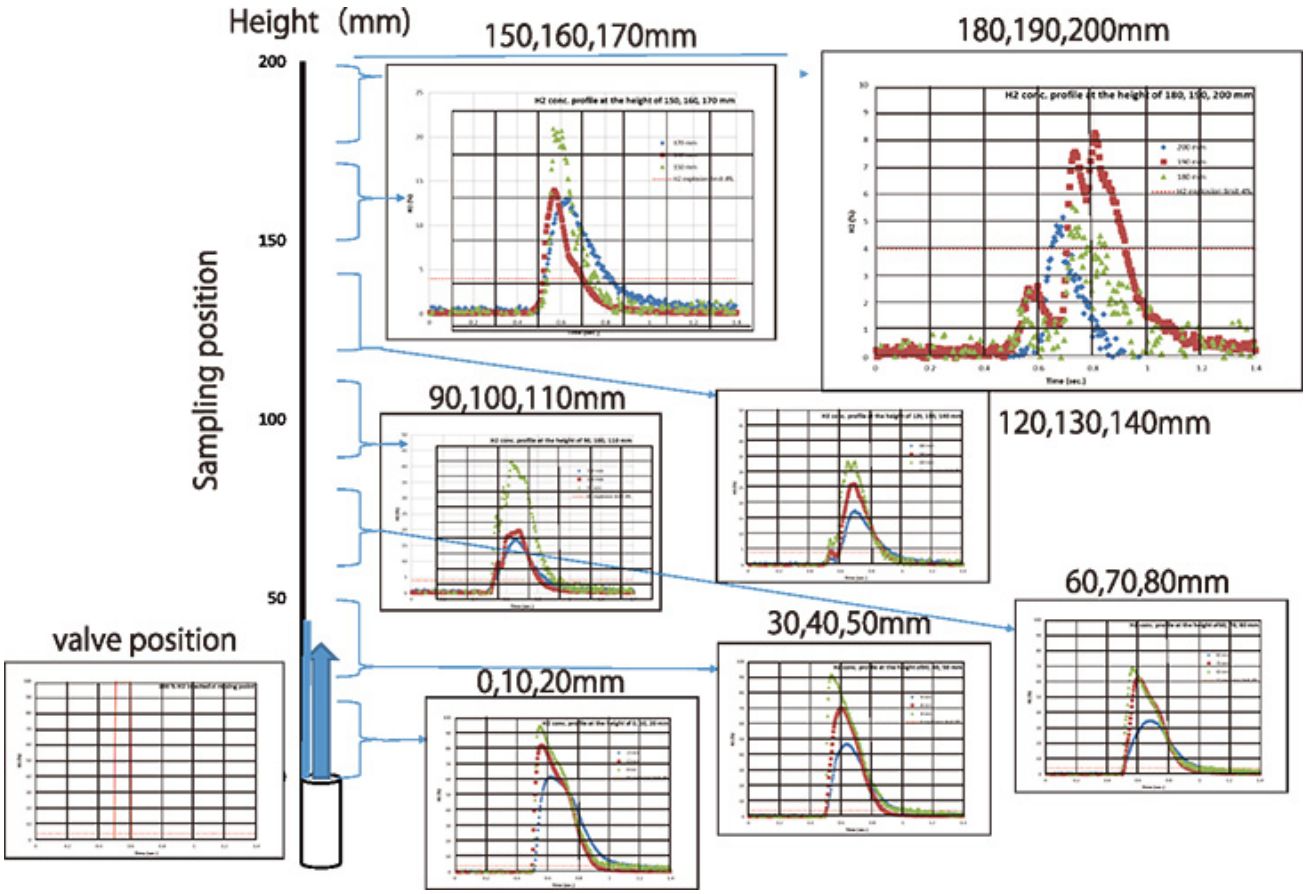

Fig.11 H2 Concentration profiles at the Spike head by changing X-Y MS gas sampling positions

; Preset H_2 concentration of 4%

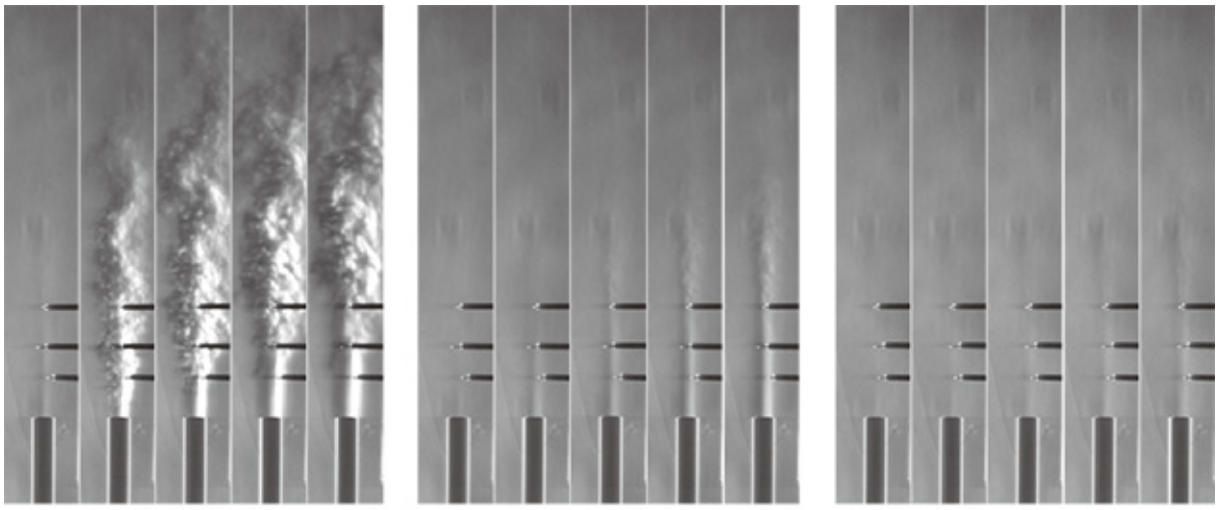

Fig.12 H_2 Injection Period 1,10,100ms into Air Flow (H_2 was introduced at the ratio of 4% in Air flow by the system shown in Fig. 3, Each Flame taken by 5ms intervals)

By comparison of Fig.11& Fig.12, we easily realized the 3D Hydrogen concentration profiles. The outside shape of clouds in Fig.12 and inside concentration of H_2 in Fig.11. MS monitored inside concentration of clouds and the Schlieren Photograph showed the outside appearance of upstream turbulence. The result showed that MS has more and linear sensitivity toward low to high concentration of H_2 than the Schlieren Photograph. In Fig.12, we recognized the out-coming cloud images of 100ms but for 10 and 1ms. In Fig 8&9, MS data showed the out-coming cloud precisely. The Schlieren Photograph showed the outside appearance of cloud and large volume of H_2 which created more density changes in space. The Schlieren Photograph had no sensitivity towards air movement. Typical flow pattern was shown in Fig.12 in 100ms H_2 purge experiment. The cloud front goes upward by waving a bit but forming main updraft axis. MS data showed in all position inside updraft, all shapes of the spike heads are similar and the head concentration has higher than tail.

3-4 Real Time Monitoring of combustion (4%,100ms)

Combustion tests were carried out and photos by high speed camera showed explosion of H_2 in pre-set of 4% and purge of 100ms. When the head of cloud approached to the tungsten wires with high voltages, the severe explosion took place by the head at 37ms. The explosion spread up first at 71ms and propagated upward and downstream with turbulence at 81-89ms. The combustion continued with constant updraft. The most turbulent premixed explosion took place above and the premixed flame propagation continued with cone inside at 89-275ms .

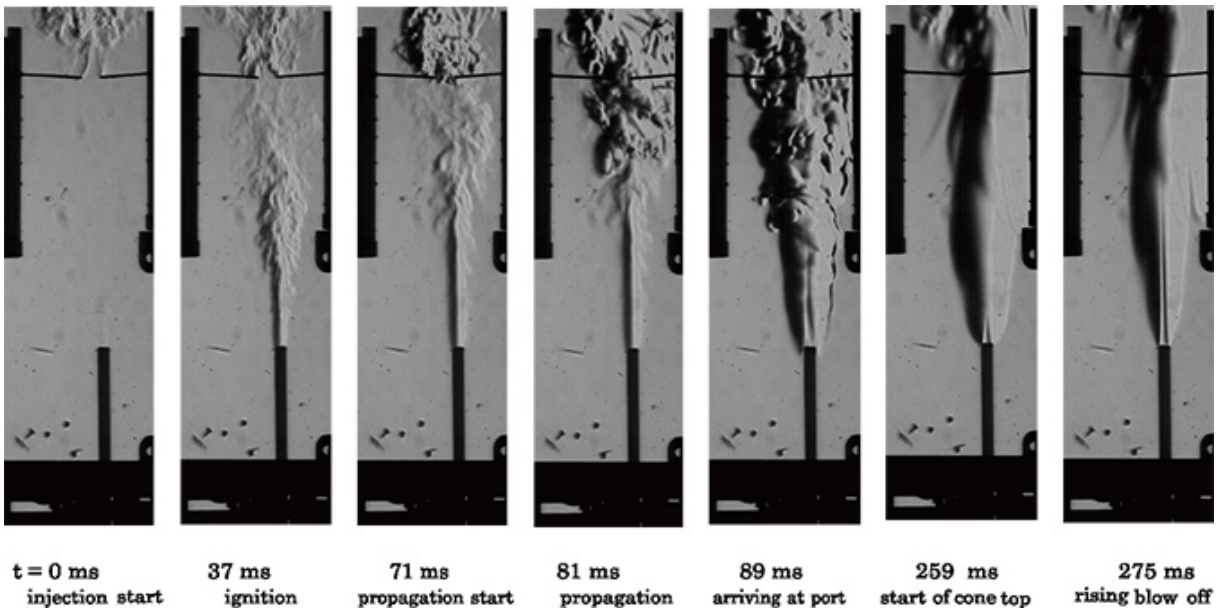

Fig.13 The Schlieren Photograph of Combustion 4% H_2,100ms in Air (H_2 was introduced at the ratio of 4% ,100ms,in Air flow by the system shown in Fig. 3 interval of 5ms)

3-5 Real Time Monitoring of combustion (4%,1ms)

The same experiment but pre-set H_2 concentration of 4% and purge 1ms took place. This Schlieren Photograph of explosion and combustion showed the existence of the higher concentration of H_2. The head of spike kept the higher concentration of H_2 to lead the explosion. In this phenomena the cone showed up in the persistent updraft to demonstrate the existence of the premixed flame.

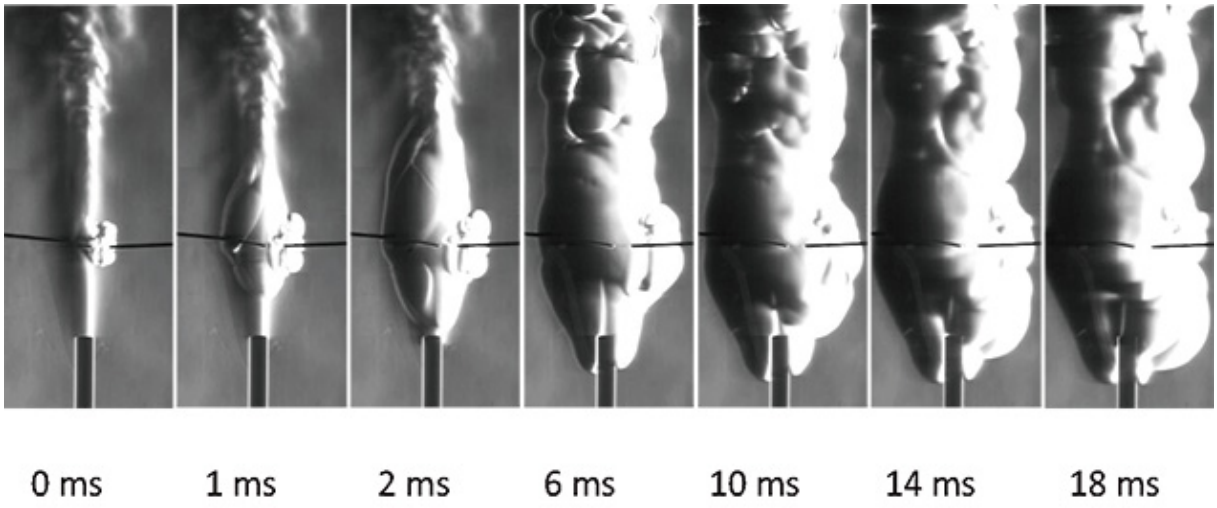

Fig. 14 The Schlieren Photograph of Combustion 4% H_2 in Air (H_2 was introduced at the ratio of 4% in Air flow by the system shown in Fig. 3)

3-6 Hydrogen Spike Head by FCV Exhaust Gas

The Schematic diagram of fuel cell is shown in Fig17. In Typical FCV, H_2 is inroduced from the pressure tank to fuel cell by decreasing pressure. H_2 purge into fuel cell through ejector. H_2 flow through hundreds of membrane with anode electrodes and come out from fuel cell to recirculate pass. The exhaust gas from typical FCV contains various gases including H_2. Hydrogen gas concentration of Typical FCV Exhaust Gas was monitored by Sx by fitting sampling cappilary at the discharging point of the center of exhaust pipe exit with small protector plate toward water droplet to come in Fig.15,16,17. The hydrogen measurement was conducted by 2-10ms interval

when FCV was parked and while idling position. So every 2ms the hydrogen concentration was taken by time. Sx data were shown in Fig. 18 and 19. In Fig.19. About every 60 second, H2 spike came out from exhaust gas as spike. The width of spike was about 1-2 second of time. By spike analysis shown in Fig. 19, one spike peak was composed of 2 or 6 spikes within 1 second and shape of spike reflecting structure of Fuel Cell stack structures. These data was taken in very short millisecond measurement and under the existence of water. These spike heads spectra analysis is pretty important to maintain the optimized operation of fuel cell and cut H_2 consumption.

Fig15 Typical FCV

Fig16 Point of Exit of FCV Exhaust System

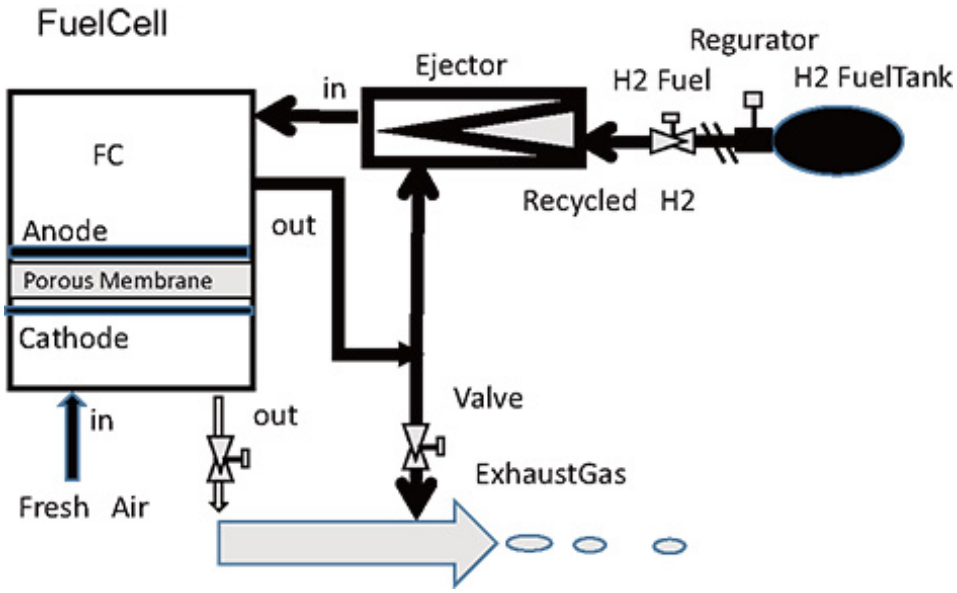

Fig.17 Schematic Diagram of Fuel Cell System in FCV

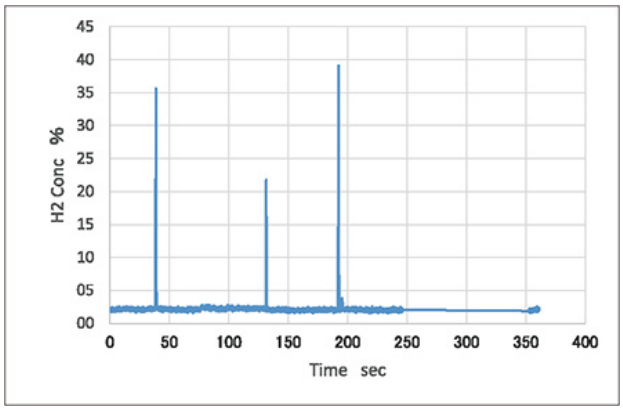

Fig. 18 H_2 H_2 Concentration Spectra at the point of Exit of FCV Exhaust System in seconds

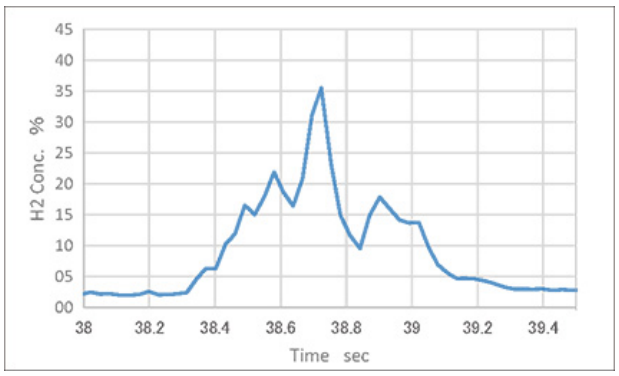

Fig.19 Spectrum of Hydrogen Concentration in FCV Exhaust Gas

4 Conclusion

Real time monitoring of Hydrogen by Sx showed the existence of the hydrogen clouds before diffusion in the mixing process. In the mixing process, pure H2 purged into air and nitrogen did not flow by forming homogeneous phase but formed heterogeneous phase like comet structure of cloud. Head and tail of cloud called the spike head and tail. The spike head analysis was carried out and demonstrated that the head portion contained high concentration of hydrogen rather than tail. Near the exit of the tube, the head of cloud, in other words, spike head contained pure hydrogen and this high concentration volume changed by injected volume. The concentration of hydrogen decreased gradually at the tail portion of this cloud.

In vertical mixing equipment, the high concentration phase of hydrogen clouds survives and moves upward by buoyancy. Once ignition equipment with high voltages applied, the explosion with premixed flame was observed by The Schlieren Photograph of Combustion 4% H_2,100ms. When mixing proceeds, the head of hydrogen cloud moves straight to form the spike head like bullet head as observed in millisecond monitored by Sx. As in the previous cylinder experiment and also the transparent plastic tube experiment, H_2 cloud moves upward rather than diffusing in all directions. In spike head H_2 stays with high concentration forming cloud and the similar spike was appeared in FCV exhaust gas in every purging actions. In basic theory of Rayleigh-Taylor instability, if the two gas phases instead of the two fluids with different densities was considered, the spike head might be came out as a cloud which

we observed by Sx. The instability of the plane interface between the two fluids, when it occurs, is called the Rayleigh-Taylor instability"

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