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Mixture distribution measurement using laser induced breakdown spectroscopy in hydrogen direct injection stratified charge

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Abstract

Reduction in cooling loss due to the heat transfer from burning gas to the combustion chamber wall is very important for improving the thermal efficiency in hydrogen engines. The previous research has shown that the direct injection stratified charge can be a technique to reduce the cooling loss and improve thermal efficiency in hydrogen combustion. For effective reductions in cooling loss by the stratified charge, it is very important to know the relation between the fuel injection conditions and mixture distribution. The current research employs the laser induced breakdown spectroscopy as a method to measure the hydrogen concentration distribution in the direct injection stratified charge. Measurement of instantaneous local equivalence ratio by the method clears the characteristics of mixture formation in hydrogen direct injection stratified charge. This research also tries to actively control the mixture distribution using a split fuel injection.

Key Words: Hydrogen Engine, Laser Induced Breakdown Spectroscopy, Stratified Charge, Mixture Distribution, Cooling Loss

1. INTRODUCTION

Hydrogen can be a clean and renewable alternative to fossil fuels. Hydrogen was first suggested as a fuel for internal combustion engines in the 1920s [1] and can be readily used in spark ignition engines [2-3]. Hydrogen internal combustion engine and proton exchange membrane type fuel cell are expected as clean power sources for transportation vehicles in the sustainable society utilizing hydrogen energy. Compared with fuel cell, hydrogen engine has the important advantages of high power density, high durability, high flexibility to impure fuels, and low cost. Only one disadvantage of hydrogen engine is the thermal efficiency, and it is very important to improve thermal efficiency of hydrogen engine. However, hydrogen has unique combustion characteristics that influence the thermal efficiency of internal combustion engines. Particularly, hydrogen has a higher burning velocity, a shorter quenching distance, and a higher thermal conductivity than conventional hydrocarbon fuels [4]. These characteristics of hydrogen exert a strong influence on the degree of constant volume and cooling loss during combustion, both of which are major factors in the thermal efficiency of internal combustion engines. The degree of constant volume indicates how close the apparent heat release is to the constant volume heat input at the top dead center, which is assumed in the Otto cycle. The cooling loss is caused by heat transfer from burning gas to the cylinder walls. The effects of operating parameters such as ignition timing, excess air

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ratio, engine speed, and coolant temperature on thermal efficiency and heat transfer in hydrogen engines have been reported in detail [5-7]. It has been shown that the heat transfer from the burning gas to the combustion chamber walls is greater with hydrogen than with conventional hydrocarbon fuel [5]. **Figure 1** shows cooling loss fraction, degree of constant volume, and indicated thermal efficiency of a spark ignition engine fueled with hydrogen and methane at a throttled stoichiometric mixture condition. Higher degree of constant volume can be achieved with hydrogen owing to its higher burning velocity than methane. However, thermal efficiency is lower with hydrogen than with methane due to the high cooling loss fraction in this condition. This signifies that the cooling loss by the heat transfer to the walls is a key factor that could be an obstacle to the thermal efficiency improvement in hydrogen combustion engines.

Operating a hydrogen engine with leaner fuel-air mixtures or retarded ignition timings is effective to reduce the cooling loss, because the leaner mixtures burn at lower temperatures and the retarded combustion phase by retarded ignition timings decreases the maximum in-cylinder temperature. The retarded combustion phase also shortens the time for burned gas and the walls in contact. However these also reduce the degree of constant volume. The leaner mixture leads to slower heat release, and the retarded ignition timing directly retards the heat release phase after top dead center. Because of this the effect in thermal efficiency improvement is limited [5]. Increasing engine speed can also reduces the cooling loss per cycle because it increases the wall temperature due to the increased heat input cycle per time [7]. The increased wall temperatures reduce the heat transfer to the walls because of the decreased differences between gas and wall temperatures. The reduction in contact time for the burned gas and the walls by increasing engine speed may also has some effect to reduce the heat transfer. However, the increased engine speed also decreases the degree of constant volume and sometimes spoils the thermal efficiency improvement effect. Therefore, a method to reduce the cooling loss without deteriorating the degree of constant volume is necessary for improving the thermal efficiency in hydrogen engines.

Direct injection stratified charge which directly injects fuel into the cylinder to locate the ignitable fuel-air mixture with a sufficient equivalence ratio around the spark plug electrode is used at lean overall mixture conditions in some types of gasoline engine [8]. The stratified charge is known to suppress the knocking which occurs when the fuel-air mixture near walls is compressed by rapid expansion of burned gas and ignites spontaneously [9]. This knock suppression is achieved because the mixture adjacent to the walls can be much leaner by the stratified charge. Such presence of leaner mixture implies that burning gas adjacent to the walls is at a lower temperature and the heat transfer to the walls is decreased. A previous report showed this cooling loss reduction effect by direct injection stratified charge with hydrogen in a constant volume combustion chamber [10]. Compared to homogeneous combustion the stratified combustion reduces the cooling loss without deteriorating the degree of constant volume, and a cycle analysis shows an effective improvement in thermal efficiency [11]. To maximize the cooling loss reduction effect by the stratified charge, it is crucial to know the relation between fuel injection conditions and mixture formation.

The laser-induced breakdown spectroscopy (LIBS) can directly measure atomic species over a wide range of mixture fractions of hydrocarbons in air [12-13]. A high-power pulse laser beam is focused at a measuring point in the fuel-air mixture and generates spark (breakdown). The resulting plasma is at temperature of 10000-25000K [14], and this is high enough to dissociate molecules into their constituent atoms, and to excite the electrons in neutral atoms and the ions in plasma from the ground state into the excited electronic states. As the plasma cools, excited electrons and ions relax back into their ground states, emitting light at characteristic atom frequencies. The atoms can be identified using atomic emission lines and can be quantified by measuring the intensity of the emission lines. The method can determine instantaneous local equivalence ratio from the spectra obtained from a single shot of the laser, and has a high spatial resolution on the order of 1mm.

This current report analyzes the influence of hydrogen injection conditions on the jet penetration and

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mixture distribution in a constant volume chamber by measuring the instantaneous local equivalence ratio using the laser-induced breakdown spectroscopy. This study also tries to actively control the stratified mixture distribution by splitting the fuel injection in two stages.

2. EXPERIMENTS

The mixture distribution with hydrogen direct injection was measured using the laser induced breakdown spectroscopy (LIBS) in a constant volume combustion chamber. **Figure 2** shows the schematic diagram of experimental system for measuring hydrogen jet. The chamber has a cylindrical shape and a volume of 227cm^3 (diameter: 85mm, width: 40mm). Before measuring mixture distribution with hydrogen jet, the correlation between fuel-air mixing ratio and atomic emissions was calibrated. The calibration curve was obtained in the following procedure. The chamber was filled with hydrogen-nitrogen mixture at a preset ratio, and the mixture was stirred with a fan driven by an electric motor. The light beam from Nd:YAG laser (Luminonics SHY750, fundamental wavelength: 1064nm, average laser energy: 80mJ/pulse) was focused in the chamber using a fused silica convex lens (f=70mm). Atomic emissions from the focal point was led to a spectrometer (JASCO CT25C) coupled with a CCD multi-channel detector (Hamamatsu Photonics PMA50) to analyze spectra. The devices were synchronized with TTL signals from a timing generator (Ascom Time98). In the hydrogen jet measurement, hydrogen was directly injected into the chamber filled with nitrogen at 0.5MPa. The injection was achieved with an electromagnetic fuel injector with a single nozzle (nozzle hole diameter: 2mm) controlled with an injector driver unit and a pulse generator (Tektronix AFG3000). The fuel feed pressure to the injector was set at 5MPa. The injector was installed on the top of the chamber and the direction of jet was perpendicular downward. In the same way as the calibration, the laser beam was led into the chamber and local concentration of hydrogen was measured using the atomic emissions. The experiments were performed ten times for each experimental condition and obtained data were averaged for maintaining good reproducibility of the results.

3. RESULTS AND DISCUSSIONS

3.1. Calibration of equivalence ratio and atomic emission intensity ratio

Figure 3 shows example spectra of atomic emissions from hydrogen-nitrogen mixture measured with LIBS. In this research, spectra were recorded in a wavelength range from 600nm to 900nm. Four strong atomic emission peaks are observed in this wavelength range. The peak at 656nm is from hydrogen, while other three peaks at 745nm, 823nm, and 871nm are from nitrogen [15]. For the nitrogen atomic emission, the peak at 871nm is the strongest of the three and this research chose it to compare with that of hydrogen for acquiring nitrogen/hydrogen fraction with high S/N ratios. Molecules in the measured volume are dissociated to the state of plasma by the high energy laser beam. The intensity of plasma emissions is weaker than those of atomic emissions, however as shown in **Figure 3**, continuous plasma emissions are observed over a wide range of wavelength. Because this research is to measure the mixture distribution using the atomic emissions, the plasma emission can be an error factor especially when the atomic emissions are relatively weak. The atomic emissions can be weak when the mixture is too rich or too lean. For this reason, emission peak values for hydrogen at 656nm and nitrogen at 871nm were measured with a baseline of the plasma emission intensity. Comparing the intensity peaks of atomic emissions from hydrogen and nitrogen gives local hydrogen concentration in the measured volume. **Figure 4** shows the relation between the equivalence ratio and the atomic emission intensity ratio of hydrogen and nitrogen. The equivalence ratio shown in the figure was calculated from the volume ratio of hydrogen and nitrogen supplied to the chamber with considering the nitrogen as air.

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3.2. Comparison of hydrogen and methane jets

Mixture formation characteristics with hydrogen jet and methane jet were compared. **Figure 5** shows the molecular ratio of hydrogen and nitrogen in the measured volume when hydrogen was injected into the chamber with the injection period of 1000μs. The measuring point with laser beam focus was scanned from nozzle tip to the jet direction. The parameter z in **Figure 5** indicates the distance from injection nozzle tip to measuring point. **Figure 6** shows experimental results when methane was injected in the same way as the experiments for **Figure 5**. The injection volume of methane was the same as the experiments with hydrogen. With the same injection amount, the injection period for methane is 2500μs. Because the viscosity of methane is higher than that of hydrogen, the injection period is longer for methane than for hydrogen.

As the measuring point is moved to downstream, the peak concentration decreases more quickly with hydrogen than with methane. This indicates that hydrogen diffuses and mixes with ambient nitrogen quickly, which can be attributed to a high diffusivity of hydrogen. On the other hand, peak concentrations at z=15mm and Z=25mm points are higher with hydrogen than with methane. Because injection period is shorter with hydrogen than with methane, hydrogen jet can be more compressed than methane jet. This may be a reason for the higher peak concentration with hydrogen. On the other hand, decrease in concentration from the peak is milder with methane than with hydrogen. This can be because diffusion coefficient of methane is smaller than that of hydrogen. In downstream, a high concentration is kept longer with methane than with hydrogen. It can be caused by differences in diffusion property and jet momentum. Therefore, the use of hydrogen, which diffuses more quickly than hydrocarbons, for direct injection stratified charge needs more efforts to locate and reside a desirable equivalence ratio around spark plug.

3.3. Axial distribution of hydrogen with split injection

Figure 7 (a) shows hydrogen concentration distribution in axial direction by single injection with 1000μs injection period. The vertical axis is the equivalence ratio calculated from molecular concentrations of hydrogen and nitrogen. **Figure 7 (b), (c), and (d)** shows the results with split injections dividing the injection quantity into 3:7, 5:5, and 7:3 respectively. The interval between first and second injections was set at 1000μs. The total hydrogen injection quantity with split injection is the same as single injection.

With single injection the injected hydrogen moves quickly from z=15mm to z=35mm. Equivalence ratio at an axial position decreases quickly from the peak showing a rapid diffusion of hydrogen. On the other hand, with split injection, the peak equivalence ratios of second jet tend to be higher than that of first jet even. This is because the second jet was injected into hydrogen mixture already formed by first jet. The presence of ambient hydrogen elevated the equivalence ratio. However, the peak equivalence ratio with split injection tends to be lower than that with single injection. In spite of this, split injections maintain ignitable equivalence ratio, 0.5 for example, longer than single injection. Especially at z=25mm, the periods with split injections is very longer than single injection. This trend is caused by changes in the jet penetration characteristics by the split injection.

Figure 8 shows schlieren images of the hydrogen jets recorded by a memory type high-speed video camera (Photoron Fastcam-ultima-RGB) using a Xenon lamp. Time-series tip penetrations of hydrogen jets are also shown in **Figure 9**. The figures show that tip penetrations by split injections are weaker than that by single injection. The tip penetrations show that the second jet reaches the slowed first jet, and this can be the reason for the increase in equivalence ratio observed in **Figure 7**. With 5:5 injection for example, the first jet stalls easily as shown in **Figure 9** and the arrival of second jet effectively maintains

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an ignitable equivalence ratio longer than single injection as shown in **Figure 7**.

3.4. Radial distribution of hydrogen with split injection

Figure 10 shows hydrogen concentration distributions in radial direction with the split injections. The measurements were done at an axial distance of z=20mm from injector tip and at a timing of 500μs after the end of injection. The hydrogen jet by single injection spreads wider into the radial direction than that by split injections. This makes the equivalence ratio on the nozzle axis lower. The equivalence ratio with the 7:3 injection is lower than that with single jet over the wide range of radial distance at the axial distance and the timing set here. This suggests that hydrogen is more widely distributed in the chamber with 7:3 injection than with single injection. On the other hand, jet by split injection with 3:7 fuel ratio tends to concentrate on the center, and this effectively avoids the diffusion of hydrogen to the radial direction. Above trends signify that the split injection can actively control the mixture distribution with adjusting the split fuel ratio. The split injection with proper settings can form and keep a high equivalence ratio mixture at a desirable location such as spark plug by weakening the jet penetration and also by the interaction of first and second jets. The hydrogen direct injection stratified charge with split injection can be an effective technique to achieve desirable mixture distribution for establishing the cooling loss reduction and thermal efficiency improvement in hydrogen engines.

4. CONCLUSIONS

The results derived by this research may be summarized as follows:

(1) The spatial distribution of hydrogen in the direct injection stratified charge can be measured with the laser induced breakdown spectroscopy. Instantaneous local equivalence ratios in a constant volume chamber were measured with intensity ratios of atomic emissions of hydrogen and nitrogen at 656nm and 871nm in this study.

(2) While hydrogen diffuses to axial and radial directions more quickly than methane with single injections, a split fuel injection in two stages can control the diffusion of hydrogen. Split injection with 7:3 fuel ratio tended to promote diffusion to wider region and that with 3:7 fuel ratio tended to weaken the jet penetration and the diffusion to the radial direction simultaneously.

(3) The split injection of hydrogen with proper settings can locate high equivalence ratio mixtures at a desirable position such as spark plug and slow the diffusion. This may be effective to control spatial distribution of mixture with direct injection stratified charge for achieving cooling loss reduction and thermal efficiency improvement in hydrogen combustion engines.

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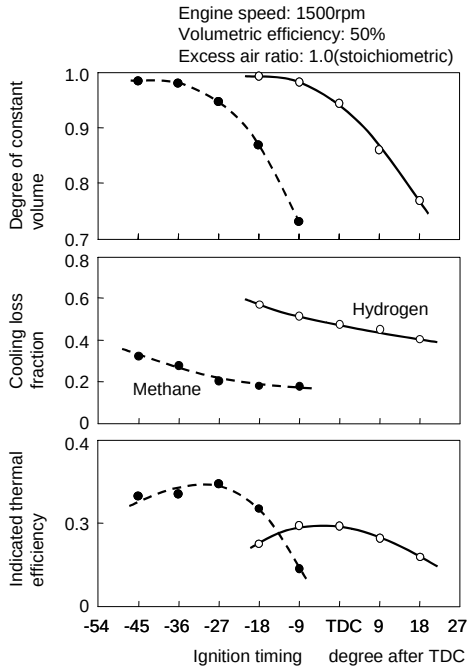

Fig.1 Cooling loss fraction, degree of constant volume, and indicated thermal efficiency for hydrogen and methane stoichiometric combustion [5].

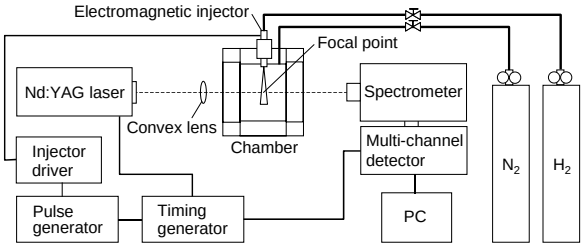

Fig.2 Experimental system for measuring equivalence ratio distribution with hydrogen direct injection using laser-induced breakdown spectroscopy.

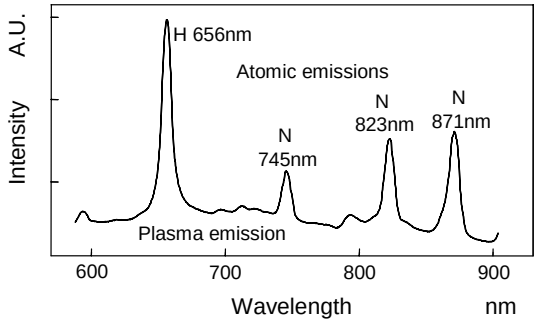

Fig.3 Atomic and plasma emissions from a hydrogen-nitrogen mixture using laser-induced breakdown spectroscopy.

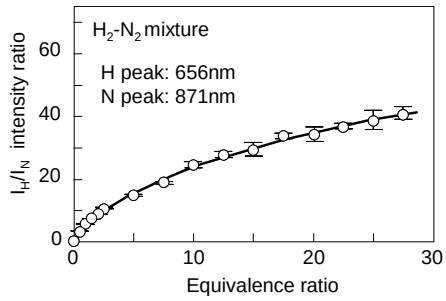

Fig.4 Calibration curve for equivalence ratio and intensity ratio of atomic emissions from hydrogen at 656nm and nitrogen at 871nm.

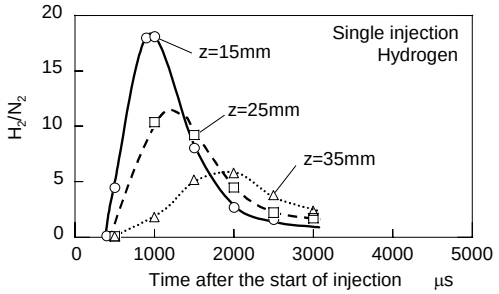

Fig.5 Time-series distribution of hydrogen on injection axis at different distances from injector nozzle tip with single injection.

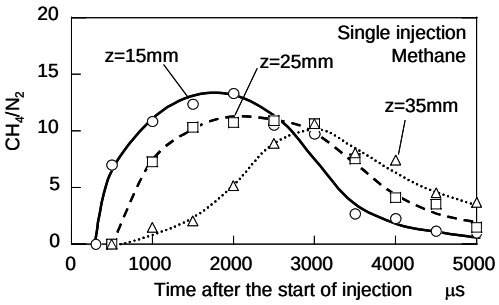

Fig.6 Time-series distribution of methane on injection axis at different distances from injector nozzle tip with single injection.

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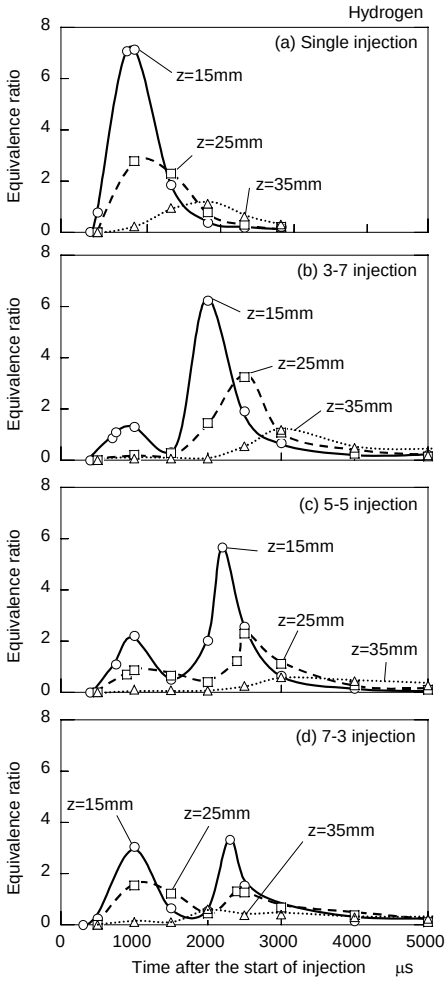

Fig.7 Time-series distribution of equivalence ratio on injection axis at different distances from injector nozzle tip with single injection and three cases of split injections.

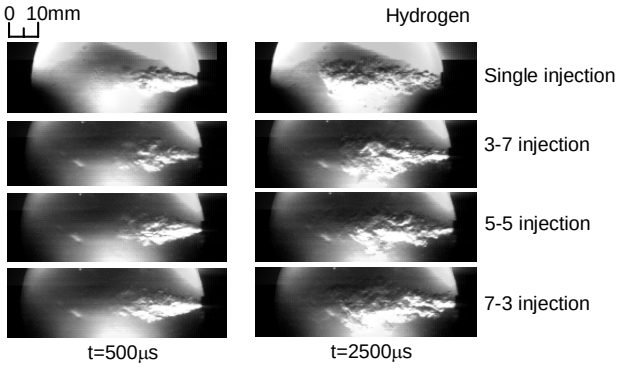

Fig.8 Schlieren images of hydrogen jets with single injection and three cases of split injections.

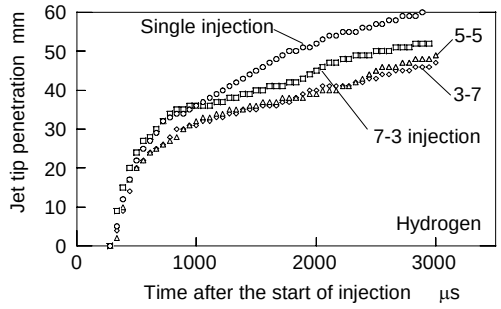

Fig.9 Time-series hydrogen jet tip penetration with single injection and three cases of split injections.

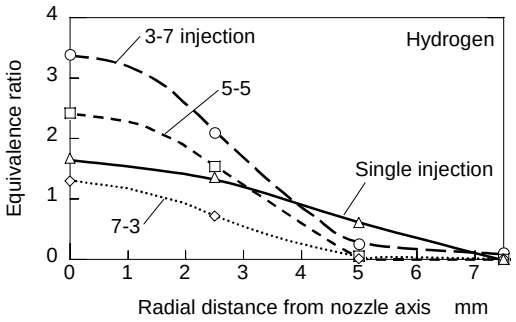

Fig.10 Radial distribution of equivalence ratio with single and split injections at an axial distance of z=20 mm from injector nozzle tip at 500μs after the end of injection.

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