An Experimental Study on Mechanism of Self-ignition of High-Pressure Hydrogen

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ABSTRACT

In the present work, self-ignition of high-pressure hydrogen released to atmospheric air through a diaphragm has been visualized under various test conditions. The experimental results show that a cylindrical flame is generated in the test tube after the self-ignition and that it continues to travel to the ambient. The present results suggest that the hydrogen which jets through the rupturing diaphragm is mixed with the heated air near the tube wall. The self-ignition event originates from this mixing, which is strongly dependent on strength of the incident shock wave generated at the diaphragm rupture. As a result, the position of the self-ignition shifts to downstream as the rupture pressure decreases. The cylindrical flame tends to become longer as it propagates in the downstream direction. Moreover, modified self-ignition mechanism is proposed based on the experimental observation.

1. INTRODUCTION

Recently much attention has been paid to hydrogen as clean energy carrier. Owing to its properties such as fast burning velocity, easy leakage, and wide flammability limit, it is well-known that hydrogen should be treated carefully in use. Nowadays operation of fuel-cell vehicles has already started, and pressure of hydrogen charged in the storage reaches 70 MPa, because hydrogen is in gas state under a room temperature. It is the pressing need of the hour to examine the characteristic of high-pressure hydrogen.

The first report on self-ignition of high-pressure hydrogen was made by Wolanski et al. in 1972 (1). Dryer et al. conducted the hydrogen release test by using rupture of a diaphragm which separates highpressure hydrogen gas and atmosphere in a test tube with circular cross-section, so that the selfignition phenomenon was duplicated (2). In the case of rectangular tubes, visualization images of the self-ignition were captured, and position of the initial shock wave, hydrogen gas jet, and the flame was observed (3). In these papers, the model of self-ignition mechanism is proposed on the basis of multidimensional incident shock waves generated at the diaphragm rupture and the reflected shock waves, which cause self-ignition in the boundary layer near the tube wall and at the center axis of the tube (2, 4). In their model, mixing of the heated air behind the shock waves and released high-pressure hydrogen proceeds so that the whole cross-section of the tube is covered with a hydrogen-air mixture. The reaction causes a flame in the boundary layer and a vortex ring appears in the core region in the tube and then reaction occurs also at this ring to generate a flame propagating in the downstream direction. These flame are merged and discharged into atmosphere from the tube end. This propagation to outside was confirmed in various experiments (5-7). The above mentioned scenario was supported by numerical simulation in several papers (8-10). In addition, it is well known that the tube length affects the critical pressure of hydrogen with which the self-ignition event is caused. Namely the longer tube leads to lower critical pressure (10-12).

Numerous attempts have been made experimentally and numerically to unravel the self-ignition phenomena of high-pressure hydrogen. However, to the authors best knowledge, the self-ignition mechanism in cylindrical tubes has not been yet verified by experimental results and no studies have ever tried to visualize self-ignition process. This paper is intended to provide the self-ignition mechanism based on visualization of self-emission in a cylindrical tube.

2. EXPERIMENTAL SETUP

Figure 1 indicates that a schematic of experimental apparatus. In operation, the test tube and the damping section are initially charged with dry air at an atmospheric pressure. Hydrogen is then charged into the storage section until a diaphragm ruptures and released into the test tube. The rupture pressure of a diaphragm, which separates the test tube from the storage section, is measured with a high-pressure gauge. The used diaphragms are made of aluminum, and their thickness is 0.4 mm. The diaphragms are scored in cruciform to rupture at a desired pressure of 5.0 MPa to 10.0 MPa in the present study. The score on the diaphragm has fixed length of 11 mm and width of 1 mm. The depth of score is measured by a surface roughness tester (Mitutoyo SJ-201) before each test, so that rupture pressure can be controlled precisely, because the rupture pressure is function of the depth of score (13). In the present work, the reproducibility of the rupture condition of the diaphragms has been well corroborated by shock speed measurement.

The test tube made of Plexiglas for visualization has an internal diameter of 10 mm and a length of 700 mm. In order to measure shock speed, two laser beams transmit perpendicularly through the test tube. Intensity of the transmitted laser beam are measured with photomultiplier tubes for detection of arrival of the shock wave. Then shock speed is calculated from time interval of the shock arrival between two adjacent locations of the laser beams. For this purpose a He-Ne laser (MELLES GRIOT 05-LHR-151) and photomultiplier tubes (HAMAMATSU H7827-012 and E7718-01) are prepared.



Figure 1. Schematic of experimental apparatus.

Sequence images of self-ignition and the subsequent flame propagation are obtained with a high speed camera (nac MEMRECAMfx K4). Flame image in the tube cross-section is also observed through the observation window fixed at the end flange of damping section, as shown in Fig. 1. In addition, an ICCD camera (HAMAMATSU C8484-05G) is arranged adjacent to the high speed camera. Trigger timing of the ICCD camera is adjusted using a delay generator (Stanford Research System DG535) to obtain an image of the self-emission at a desired time.

3. RESULT AND DISCUSSION

3.1 Self-ignition

Figure 2 shows typical images of the self-ignition process for higher rupture pressure and lower. The vertical line shows elapsed time after the self-ignition is first observed, and the horizontal one distance from the diaphragm. In Fig. 2 (a), it is shown that the self-emission is detected at 240 mm at time zero, while the self-ignition occurs at 460 mm in Fig. 2 (b). These images indicate that the position of the self-ignition shifts to downstream as the rupture pressure decreases. This agrees well with the previous

results which clarify effects of the tube length and the rupture pressure on self-ignition of released hydrogen (10-12). Even slight reduction of approximately 1.8 MPa (from 8.9 MPa to 7.1 MPa) in the rupture pressure makes almost twice the distance of the self-ignition phenomena.



Figure 2. Direct images of the self-ignition process in the test section observed from observation window A with high speed camera. The rupture pressure and shock speed: (a) 8.9 MPa, 1600 m/s, (b) 7.1 MPa, 1500 m/s.

Figure 2 also demonstrates that the flame tends to become longer as it propagates in the downstream direction. The velocity of the central part of the flame is estimated to be 1300 m/s from Fig. 2 (a), while the head portion of the flame moves at 1400 m/s and the tail portion at 1200 m/s. This central velocity is generally consistent with that of the contact surface, which is calculated from the measured shock speed. This suggest that the central part of the flame moves at the same speed with the main air flow behind the shock wave and that the head and tail portion of the flame propagate at a relative speed of 100 m/s in the downstream and upstream direction, respectively. This relationship between the contact surface and the flame movement is the same case with Fig. 2 (b), in which the flame has a range of speeds from 900 m/s to 1300 m/s. Higher rupture pressure generates stronger shock wave, making movement of the contact surface faster. Consequently, swift movement of the flame is attained in the case of higher rupture pressure. It is obvious that there exists difference in the relative speed of the flame to the main air between Fig. 2 (a) and (b). While at 250 µs, the flame length in Fig. 2 (a) is about 90 mm, this length is achieved at 100 µs in Fig. 2(b).

As for intensity of the self-emission, it seems that luminosity of the flame is higher in the Fig. 2 (a) than in Fig. 2 (b). This is probably due to the fact that the self-emission is a visible-light image. Time for occurrence of the self-ignition increases with decrease in the rupture pressure, causing enhancement of degree of premixing of the hydrogen jet and the surrounding air. Since the hydrogen flame has essentially ultraviolet emission of OH chemiluminescence, progress in the degree of mixing gives lower visible luminosity of the flame.

3.2 Flame shape

Typical self-emission images taken by the ICCD camera are displayed in Fig. 3, where the images (a) and (b) were obtained in the same test with Fig. 2 (a) and (b), respectively. The change of color from blue to white represents increase of brightness. It is clear that the luminous region spreads in the axial direction as the flame moves in the downstream direction. Although in Fig. 3 (b) it is difficult to discern the flame owing to its low luminosity as mentioned above, the flame has almost the same length with the third image of Fig. 3 (a).

Figure 4 exhibits typical images of the self-ignition process in the test tube observed from the downstream through the observation window fixed at the end flange of damping section. In Fig. 4 (a) self-ignition is detected at first near the lower wall in the test tube. Thereafter this self-ignition generates the ring-shaped or cylindrical flame as shown in Fig. 4 (b). This ring-shaped emission is consistent with the high-speed images in Fig 2 which shows that there are two luminous regions along the upper and lower tube wall.

Figure 5 shows typical image of self-emission obtained with ICCD camera to capture outline of the flame in more detail. The cylindrical flame tends to shrink in the downstream, mainly because of form of hydrogen jet penetrating into the main air flow behind the shock wave.



Figure 3. Direct images of the self-ignition process in the test section observed with ICCD camera. The rupture pressure and shock speed: (a) 8.9 MPa, 1600 m/s, (b) 7.1 MPa, 1500 m/s. Gating time: 1 μ s.



(a) Just after self-ignition



(b) Ring-shaped flame

Figure 4. Images of the self-ignition process in the test section observed from observation window B with high speed camera. Rupture pressure: 8.8 MPa, Shock speed: 1600 m/s, frame rate: $4x10^4$ fps.



Figure 5. Detailed image of the self-ignition process. Rupture pressure; 8.9 MPa, shock speed: 1600 m/s. Gating time: 100 ns.

3.3 Self-ignition mechanism

Figure 5 shows schematic of self-ignition mechanism of high-pressure hydrogen release into a tube by rupture of diaphragm, inferred from the present results. Incident shock wave is generated by diaphragm rupture which is gradually opened by high-pressure hydrogen. The shock wave heats air and a boundary layer is caused along the inner wall of the tube behind the shock wave. In the present experimental conditions for successful self-ignition, air temperature behind the shock wave is estimated to be more than 900 K. Moreover, temperature in the boundary layer is slightly higher than the main air stream because viscous dissipation plays a role in high speed flows. The diaphragm and opening area is enlarged to cover whole cross-sectional area of the tube. This rupture process forms the hydrogen jet which has convex end in the movement direction. Shear mixing of the hydrogen jet and the heated air in the boundary layer is enhanced so that hydrogen-air mixture satisfy the temperature condition for self-ignition. Under high pressure conditions like in the present study, self-ignition is possible if temperature more than 750 K is achieved (14). Once self-ignition occurs in this mixing zone, flame propagates inside the mixing layer between the hydrogen jet and the shock heated air so that the cylindrical flame is formed.

Higher rupture pressure generates stronger shock wave, giving higher air temperature and enhancing the mixing process. This contributes to rapid formation of the mixing zone satisfying the temperature condition of the self-ignition and flame propagation is dominated by the mixing process. On the other hand, in the case of lower rupture pressure, self-ignition needs longer time due to lower temperature of air behind the shock wave. In this case, the mixing of hydrogen jet and the shock heated air proceeds during waiting time of the self-ignition. As a consequence, the flame propagates in the premixed hydrogen-air. This is supported by the fast flame propagation and the lower luminosity as mentioned before.



(i) Higher rupture pressure





Figure 6.Mechanism of self-ignition process, (a) immediately before rupture; (b) generation of shock wave; (c) occurrence of mixing zone; (d) self-ignition at higher rupture pressure; (e) formation of cylindrical flame at higher rupture pressure; (f) propagation of the flame at higher rupture pressure; (d') extension of mixing zone without ignition at lower rupture pressure; (e') self-ignition at lower rupture pressure; (f) propagation of the flame at lower rupture pressure.

4. CONCLUSION

In the present study, the self-ignition process has been visualized to reveal self-ignition mechanism. The following conclusions have been obtained from the experimental results.

- 1. The position of the self-ignition shifts to downstream as the rupture pressure decrease.
- 2. The cylindrical flame is generated after the self-ignition and tends to become longer as it propagates in the downstream direction.
- 3. Modified self-ignition mechanism is proposed based on the experimental observation.

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