Self-Ignition of High-Pressure Hydrogen Released by Reproducible Rupture of Diaphragm

W. Kaneko, K. Hayashi, K. Ishii
Department of Mechanical Engineering, Yokohama National University, 79-5 Tokiwadai Hodogaya-ku, Yokohama, Kanagawa 240-8501, Japan

1 Introduction

In recent years, dwindling fossil fuels argue a necessity of alternative energy sources, because carbon dioxide has caused global warming. Clean hydrogen energy is a dominant candidate to solve this problem. However, on its use, hydrogen gas in a storage tank is usually compressed to high pressure owing to its low energy density. Then high-pressure hydrogen of 35 MPa or 70MPa is supplied to hydrogen stations and fuel-cell vehicles. It is well known that the high-pressure hydrogen gas can ignite spontaneously without any ignition sources, if it leaks from storage or a high pressure pipe.

In several papers the cause of the self-ignition is explained as follows [1-5]: First, a safety valve or a damaged device suddenly releases high-pressure hydrogen gas into a tube, which has various shapes and contains air in it. Release of the high-pressure hydrogen gas generates a shock wave heating surrounding air around the release area. As a result, mixing of the heated air and the released hydrogen causes self-ignition. In addition, it is reported that storage pressure and the tube length affect the self-ignition process [2-4, 6]. Namely, a longer tube and higher storage pressure give higher probability of the self-ignition.

In experiments of these papers, the self-ignition of high-pressure hydrogen, which is suddenly released from storage, is realized by rupturing a diaphragm separating the high-pressure gas from an ambient air. In these experiments the critical rupture pressures, which is minimum pressure for the self-ignition have been obtained, using various kinds of diaphragms. However, the results show that the critical pressure disperses, even if the rupture tests are conducted on the same conditions. One reason for this dispersion might be differences of the diaphragm opening process, because the diaphragm opening is not instantaneous, and requires a certain period of time. An opening time and an opening manner of the diaphragms vary under the same rupturing conditions [5], resulting in variation of shock wave formation.

In the present study, care has been taken to improve reproducibility of rupturing diaphragms and the critical pressure for the self-ignition of hydrogen has been obtained using various kinds of diaphragms. Moreover, the self-ignition process has been visualized to reveal self-ignition mechanism of the released hydrogen.

2 Experimental

Figure 1 shows a schematic of experimental apparatus. The test section and damping section are initially charged with dry air at an atmospheric pressure. Hydrogen is then charged into the storage

Correspondence to: kaneko-wataru-wt@ynu.jp

section until a diaphragµ ruptures. A pressure gauge measures the rupture pressure of a diaphragm, which separates the storage section from the test section is measured with a conventional pressure gauge. The test section is composed of six identical parts, and a length of each part is 54 mm. Each part has a channel of 10 mm in internal diameter and is equipped with a conventional pressure transducers (PCB 112A24) denoted by P1-P6 in Fig. 1. Length of the test section can be varied from 80 mm to 404 mm by combining the parts. The capacity of the storage section is sufficiently larger than that of the test section, so that expansion waves generated by rupture of diaphragm do not arrive at the damping section until the shock wave plunges into the damping section. The diaphragms used are made of aluminum, brass and copper and their thickness is 0.2 mm, 0.4 mm and 0.5 mm. The diaphragms are scored in cruciform to rupture at a desired pressure. The score on the diaphragm has fixed length of 11 mm and width of 1 mm. Its depth is changed from 30 µm to 160 µm, which corresponds to rupturing pressure of 3.0 MPa to 15.0 MPa in the present study. The depth of score is measured by a surface roughness tester (Mitutoyo SJ-201) before each test.

Two observation windows are fixed at the end flange of the storage and damping section, as shown in Fig. 1, to observe opening process of the diaphragm and the self-ignition process. A xenon lamp irradiates the diaphragm through the observation window A. Light from the xenon lamp filtering through the diaphragm during its opening process is recorded with a high speed camera (nac MEMRECAMfx K4), which is placed near the observation window B so that opening time of the diaphragm is estimated. After rupturing of the diaphragm, a shock wave is formed in the test section and propagates towards the damping section. Strength of the shock wave is measured with the pressure transducer and then a shock speed is calculated from the time interval of shock arrival between two adjacent locations of pressure measurement. The reproducibility of the diaphragm rupturing is evaluated by the opening time of the diaphragm, the rupture pressure, and the measured shock speed. Light intensity of self-emission at the self-ignition event is measured with a photomultiplier tube (HAMAMATSU H7827-012) through the observation window A, while the self-emission image is recorded with the high speed camera through the observation window B.

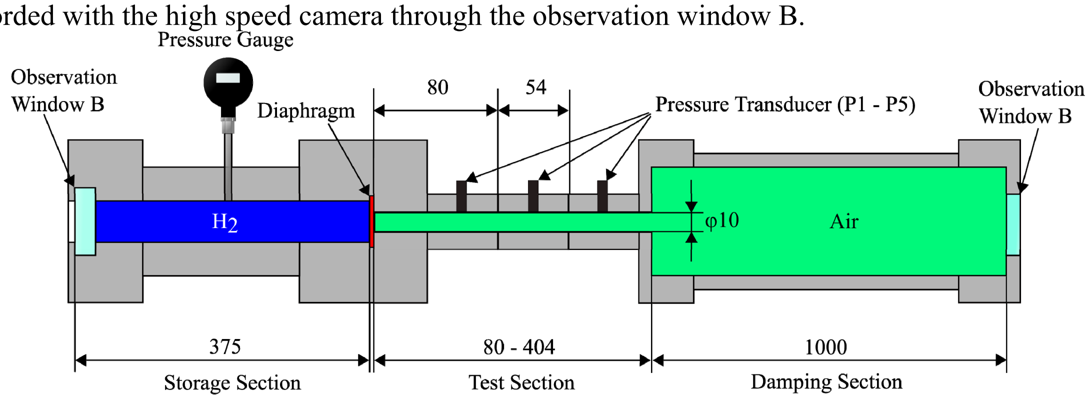

Figure 1. Schematic of shock tube.

3 Results and Discussion

3.1 Reproducibility of diaphragm rupture and critical conditions for self-ignition

Figure 2 shows the relation between the depth of score on the diaphragm and the rupture pressure. Because the surface roughness tester used in the present study has limitation in measurable ranges in the depth of score, which is also dependent on thickness of the diaphragm, various kinds of diaphragm in terms of material and thickness provide a wide ranges of the rupture pressure of 5.0 MPa to 15.0 MPa, as shown in Fig. 2. The diaphragms used are copper of 0.2 mm in thickness, brass of 0.2 mm, and aluminum of 0.4-0.5 mm. Figure 2 shows that the rupture pressure linearly increases with decrease in the depth of score. This linear relation indicates that the rupture pressure can be controlled with good reproducibility by selecting an appropriate diaphragm material and the depth of score. The copper and brass diaphragms used in the present study are thinner than the aluminum ones, so that a slight variation in the depth of score in the former diaphragms results in relatively larger change of the rupture pressure. This is the reason why the aluminum diaphragms, which is thicker than the others, exhibit better linearity in Fig. 2.

Figure 3 shows effects of the length of the test section and the rupture pressure on self-ignition of released hydrogen. It is indicated that the longer test section lead to the self-ignition, even if the rupture pressure is low. However, there is a little variance of critical rupture pressure for self-ignition under the condition that the length of the test section is the same. The reason for this variance is that there are a few diaphragms which rupture at not expected pressures. Consequently these diaphragms give different self-ignition process as compared to ordinary ones. Moreover, the diaphragms made of various kinds of metal or in thickness have different opening time, even if they ruptures at the same storage pressure.

Then this result shown in Fig. 3 is re-arranged in terms of the shock speed, as shown in Fig. 4. It is well demonstrated that the shorter test section needs the higher shock speed for self-ignition. It must be noted that the critical shock speed for self-ignition does not disperse in these results.

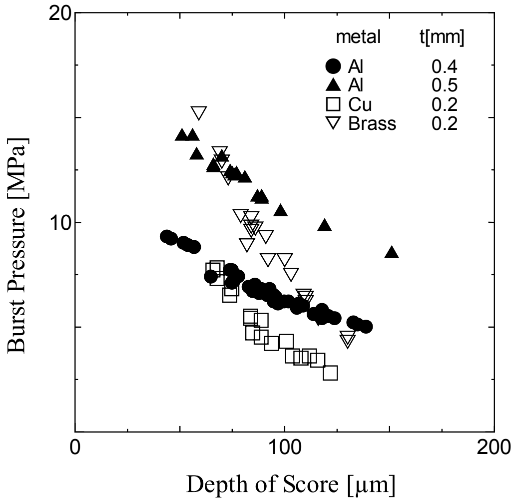

Figure 2. Relation between depth of score on diaphragm and rupture pressure.

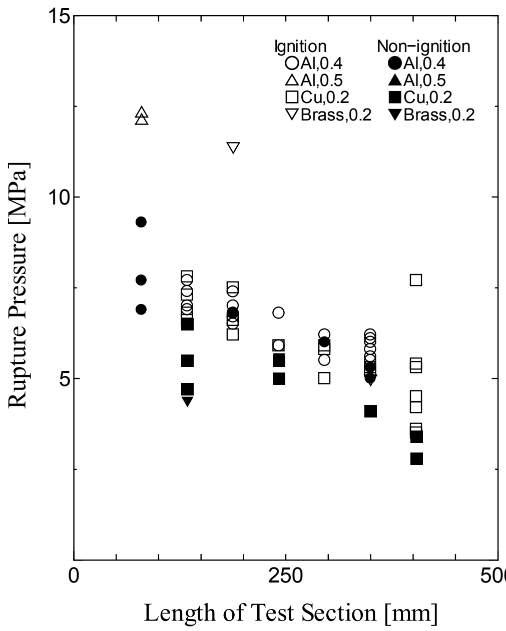

Figure 3. Effects of length of test section and rupture pressure on self-ignition of released hydrogen.

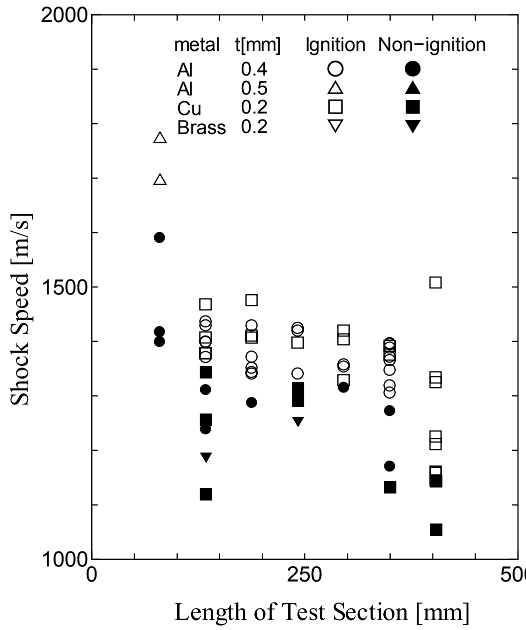

Figure 4. Effects of length of test section and shock speed on self-ignition of released hydrogen.

3.1 Visualization of self-ignition in test tube

Typical direct images of the self-ignition process are displayed in Fig. 5. Self-ignition is detected at first near the lower wall in the test section at t = 114 μs. This self-ignition causes the semi-ring shaped flame at t = 133 μs, at which another self-ignition is observed near the upper wall. At t = 152 μs two flames in the upper and lower wall develops independently, forming into a ring-shaped flame at t = 171 μs. Afterwards the ring-shaped flame travels from the test section to the damping section. In this case, the flame is often extinguished partly during traveling downstream and then totally extinguished at t = 257 μs. The shock wave constantly propagates ahead of the ring-shaped flame in the test tube and then plunges into the damping section at 171 μs.

Formation of the ring shaped flame is responsible for mixing process between the released hydrogen and the ambient air in the test section. In rupturing the diaphragm, hydrogen jets through an opening portion of the diaphragm, which generates hydrogen core near the center axis in the test section. This released hydrogen is mixed with surrounding air, which has been heated up by the shock wave. This mixing mainly proceeds near the boundary layer at the wall induced by the flow behind the shock wave. The mixture of the released hydrogen and the shock heated air, therefore, is formed near the wall, and, is ignited to form the ring-shaped flame around the inner surface of the test section after a certain ignition delay time, if the mixture satisfies the necessary conditions for self-ignition. Afterwards the flame travels with keeping its ring form, not propagating in the radial direction of the test section.

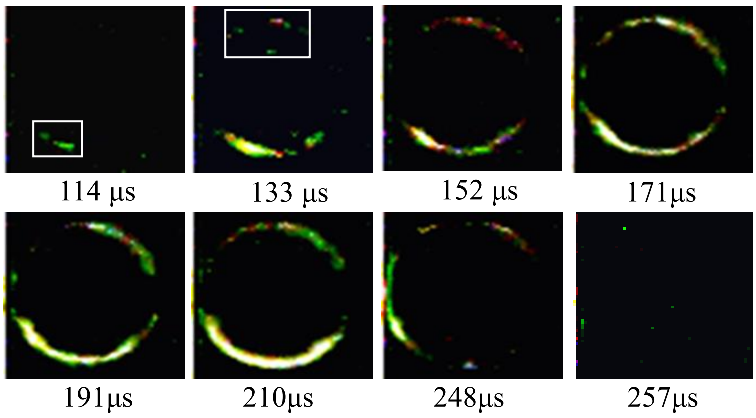

Figure 5. Direct images of the self-ignition process in the test section observed from the downstream. Shock speed is 1760 m/s and length of test section is 296 mm.

4 Summary

In the present study, care has been taken to improve reproducibility of rupturing diaphragms and the critical pressure for the self-ignition of hydrogen is obtained using various kinds of diaphragms. Moreover, the self-ignition process has been visualized to reveal self-ignition mechanism.

1. The rupture pressure can be controlled with good reproducibility by selecting an appropriate diaphragm material and the depth of score. This fact leads to little dispersion in the critical shock speed for self-ignition.
2. The shorter test section needs the higher shock speed for self-ignition.
3. Mixing process between the released hydrogen and the ambient air in the test section is responsible for formation of the ring shaped flame.

References

[1] Dryer FL et al. (2007). Spontaneous ignition of pressurized releases of hydrogen and natural gas into air. Combust. Sci. Technol. 179: 663.

[2] Oleszczak P, Wolanski P. (2010). Ignition during hydrogen release from high pressure into the atmosphere. Shock Waves. 20: 539.

[3] Bragin MV, Molkov VV. (2011). Physics of spontaneous ignition of high-pressure hydrogen release and transition to jet fire. Int. J. Hydrogen Energy. 36: 2589.

[4] Golub VV et al. (2008). Mechanisms of high-pressure hydrogen gas self-ignition in tubes. J. Loss Prevent Prov. Indus. 21: 10956.

[5] Golovastov S, Bocharnikov V. (2012). The influence of diaphragm rupture rate on spontaneous self-ignition of pressurized hydrogen: Experimental investigation. Int. J. Hydrogen Energy. 37: 10956.

[6] Mogi T et al. (2009). Self-ignition and flame propagation of high-pressure hydrogen jet during sudden discharge from a pipe. Int. J. Hydrogen Energy. 34: 5810.