EXPERIMENTAL STUDY ON AUTO-IGNITION OF HIGH PRESSURE HYDROGEN JETS COMING OUT OF TUBES OF 0.1-4.2M IN LENGTH

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ABSTRACT

Wide use of hydrogen faces significant studies to resolve hydrogen safety issues in industries worldwide. However widely acceptable safety level standards are not achieved in the present situation yet. The present paper deals with hydrogen leaks from a tube to ignite and explode in atmosphere. The experiments using a shock tube are performed to clarify the auto-ignition property of high pressure hydrogen jet spouting from a tube. In order to improve experimental repeatability and reliability, the shock tube with a plunger system is applied, where the PET diaphragm is ruptured by a needle in order to control a diaphragm burst pressure (hydrogen pressure). As a result, it becomes possible to control the diaphragm burst pressure to obtain a local minimum value. The most important result obtained in the preset study is that the minimum diaphragm burst pressure for auto-ignition is found between 1.0 and 1.2 m of tube length using a longer tube than the one used in the previous study. This minimum tube size is not found elsewhere to suggest that the tube length has a limit size for auto-ignition. Furthermore auto-ignition and Mach disk at the tube exit are observed using a high speed camera which is set at the frame speed of 1×10^5 fps when the ignited hydrogen jet is spouted out the tube.

1.0 INTRODUCTION

Hydrogen is considered as an alternative renewable source of energy, practically inexhaustible source of energy and absolutely clean as well and may replace fossil fuel. Some are interested in hydrogen to use as a new energy source. Others are interested in hydrogen from scientific and technological points of view. Power generation and transportation companies are also interested in technology and business of hydrogen infrastructure. Many governments think hydrogen technology to be a priority matter for their social and economic development. Hydrogen energy is one of the answers for the global environmental issue.

However, there are some problems when hydrogen energy safety issue is concerned. One of the serious problems is that hydrogen may lead to an accidental explosion. High pressure tank is often used to store hydrogen. It is necessary to compress hydrogen gas to store a large amount since hydrogen energy density per volume is low. However, it is well known that the accidental release of hydrogen from a high-pressure tank into air can produce a strong shock wave that heats up hydrogen and air to a high temperature to ignite hydrogen-air mixture. The auto-ignition of hydrogen in air leads to an explosion under a certain condition. From the safety point of view, hydrogen explosion problem is important in practical cases such as pressure vessels containing high-pressure hydrogen, automobile fuel cell, etc.

For this reason, hydrogen ignition studies have been performed for more than decades to clarify the mechanism of an explosion induced by high pressure hydrogen jet spouting from a hole and a tube [1–10]. Auto-ignition of hydrogen jet at the tip of the contact surface between hydrogen and air is confirmed by many studies, but the mechanism of extinction or stabilization in such cases is still under discussion. The different spouting jet conditions in these studies shows different results: i.e., Golub et al. [6], Mogi et al. [7] presented a relationship between hydrogen inlet pressure and tube length. From their results it is found that auto-ignition using a long tube occurs easier than that using a short one.

When hydrogen jet is spouted into a tube connected to air atmosphere, the shock wave yields ahead of hydrogen jet there and even in the tube. When the shock wave reaches the exit of the tube, the gas begins to expand widely due to pressure difference between hydrogen pressure and atmospheric air pressure and the vortices grow up around the tube exit, and an ignition and explosion of hydrogen is induced [10]. When high pressure hydrogen propagates in a long tube connected to atmospheric air, it is expected that an explosion may also occur in the tube. This explosion is still not sure where and how it happens in the tube. Then such ignition and explosion in tube must be studied in detail from the safety point of view.

2.0 EXPERIMENTAL SETUP

Figure 1 shows a schematic of the present experimental system. Experimental devices are composed of three sections; a driver section with a needle to rupture a diaphragm which holds high pressure hydrogen and a diaphragm burst pressure sensor (P_b), a shock tube (the extension tube in Fig.1) with five pressure sensors (P_1 through P_5), where hydrogen spouts suddenly at the burst diaphragm, and a plunger system with a coil to generate magnetic field for controlling a needle. PET film is used as the diaphragm, which thickness is from 25 to 188 μm, therefore it is possible to set any burst pressure through a calibration (Table 1). Experiments are performed up to the maximum pressure of approximately 8 MPa. The electrically polished tube is used as the shock tube with its internal diameter of 10 mm. The length of the extension tube can be changed from 0.1 to 4.2 m. The pressure sensors are set on the upper wall of the shock tube at 30 cm intervals to record pressure histories in an oscilloscope.

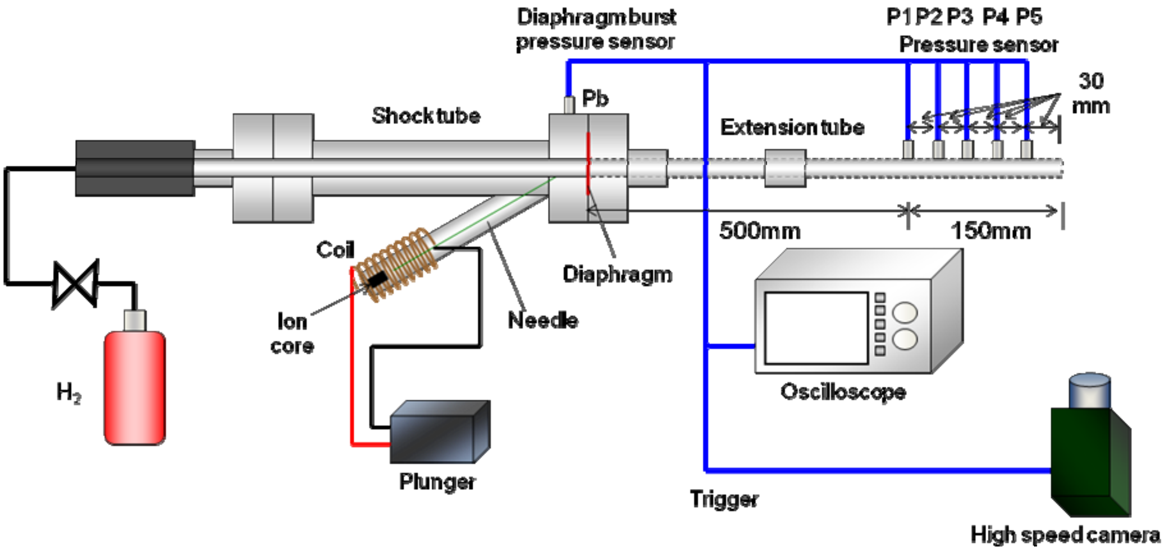

Fig. 1 Schematic of experimental system

Table 1 Relation between diaphragm thickness and initial burst pressure

Diaphragm thickness, μm	Limit burst pressure, MPa	Initial burst pressure, MPa
25	1.5	1.0
38	1.8	1.5
50	2.2	2.0
75	3.0	2.5
100	4.0	3.8
125	5.0	4.8
150	6.7	6.3
188	8.0	7.5

Hydrogen pressure is set by the diaphragm burst pressure (P_b) which is closed to the one used by the previous work done by Mogi et al. [7] where they used the so-called burst pressure that the diaphragm is ruptured naturally by the hydrogen pressure. In the previous work the diaphragm burst pressure is not controlled because we do not know exactly when the diaphragm ruptured. In the present work the rupture is done by the needle according to our decision. Hence the diaphragm burst pressure can be chosen by us.

3.0 RESULTS AND DISCUSSION

Experiments are performed for seven categories; (1) the pressure profiles for the cases of (a) auto-ignition, (b) blow-off, and (c) failed-ignition; (2) auto-ignition when the tube length (the extension tube length) is variable at a constant diaphragm burst pressure (initial hydrogen pressure to the extension tube (Fig.1)); (3) auto-ignition when the diaphragm burst pressure is variable using a constant tube length; (4) reproducibility of experiments; (5) shock velocity for varying diaphragm burst pressure; (6) auto-ignition limit for various shock tube lengths and various diaphragm burst pressures; (7) visualization of auto-ignition using a high speed camera.

3.1 Pressure profiles for three cases; auto-ignition, blow-off, and failed-ignition

(a) Auto-ignition case

The past experiments [6-7] show that auto-ignition has a strong relation with the diaphragm burst pressure (initial hydrogen pressure spouting to the extension tube) and shock tube length. The present experiments show the similar results of such relation. The pressure transducers are located at 500 mm (P1), 530 mm (P2), 560 mm (P3), 590 mm (P4), 620 mm (P5) from the diaphragm position (Fig.1). The pressure history for each sensor is not shown here, but the first pressure rises for P1 through P3 are not straight upward due to underdevelopment of shock wave. The highest pressure peak comes after the first rise of the shock wave because the shock wave goes ahead of the contact surface between hydrogen and air and is about 2.4 MPa, where the length of the tube used is 650 mm and the initial diaphragm burst pressure is 6.3 MPa.

(b) Blow-off case (extinguishment after auto-ignition)

The blow-off case implies that the auto-ignition is recognized by bright light or sound to extinguish afterwards. The pressure profiles of the blow-off cases are similar to that of auto-ignition case, but the big difference between the blow-off case and the auto-ignition case is that the maximum pressure of the blow-off case is about 0.4 MPa lower than that of auto-ignition case. This means that the pressure of the blow-off case at the contact surface between hydrogen and air is lower than that of the auto-ignition case. This low pressure gives no continuation of ignition. This case will be discussed in

Section 3.7 of visualization of auto-ignition using a high-speed camera, where the sound does not help to distinguish between auto-ignition and blow-off case.

(c) Failed-ignition case

The failed-ignition means that the auto-ignition does not happen and the head shock pressure does not go up much (Fig.2). The pressure profiles of the failed-ignition cases are a little different from that of the auto-ignition and the blow-off case: i.e., the first pressure rise is straight, but after that the pressure drops about 0.2-0.3 MPa, then increases to its peak, which is 0.4 MPa lower than that of the auto-ignition case and similar to that of the blow-off case.

In summary the auto-ignition occurs when the tube length of 650 mm and the initial diaphragm burst pressure of 6.3 Mpa are applied (this will be discussed in detail in Section 3.4). The maximum pressure rise for the auto-ignition case is 2.4 MPa, which is the pressure at the contact surface between hydrogen and air. The physics in the region between the precursor shock wave and the contact surface provides the difference between the ignition and non-ignition case; in other words, the strong head shock provides the strong flow at the contact surface between hydrogen and air with a larger vortex, but the weaker head shock does not give such strong vortex at the contact surface, which gives an extinguishment of ignition. This can be seen in the numerical simulation such as shown in Yamada et al. [9].

3.2 Experiments of auto-ignition at the constant diaphragm burst pressure when the tube length varies

Auto-ignition strongly relies on diaphragm burst pressure and tube length. Hence in this section auto-ignition is studied at the constant diaphragm burst pressure, but at the variable shock tube length. Three constant diaphragm burst pressure cases at the pressure sensor P_b are applied this time and especially the case of the diaphragm burst pressure of 4.8 MPa is discussed here. The experimental conditions such as tube length, diaphragm burst pressure, diaphragm thickness, mean shock speed, and ignition or no ignition status at the diaphragm burst pressure of 4.8 MPa are shown in Table 2 where the mean shock speed is obtained from the average speed between two pressure sensors where the pressure sensor P1 is located at 150 mm inside from the tube exit (Fig.1).

Table 2 Conditions at the diaphragm burst pressure P_b of 4.8 MPa

	Tube length, mm	Burst pressure, MPa	Thickness of diaphragm, μm	Mean shock speed, m/s	Auto-ignition or no-ignition
1	400	4.8	125	1045	×
2	1200	4.8	125	1002	○
3	1700	4.8	125	939	×
4	3200	4.8	100	780	×

Figure 2 shows the typical pressure histories on the inner tube wall for various tube lengths at the diaphragm burst pressure of 4.8 MPa, which conditions are also shown in Table 2. In general the wall pressure becomes lower and the maximum pressure decreases downstream as the tube length becomes longer due to the wall friction and expansion effect. In Fig.2 the first pressure rise at each pressure profile is the head shock and the following maximum pressure is at the contact surface between hydrogen and air as described in the section 3.1. Only the case of the tube length of 1200 mm has an auto-ignition although the case of the tube length of 400 mm has the higher maximum pressure of 2.0 MPa. The 1200 mm tube length case has the maximum pressure of near 1.5 MPa, but it keeps the average pressure of about 1.3 MPa for 0.7 ms which may provide the mean shock speed of 1002 m/s and lead to the auto-ignition (Table 2). Together with the numerical results of the hydrogen flow physics in the tube (not shown in this paper, but discussed in Yamada et al., 2011 [9]), the vortices appear between the head shock and the contact surface, which reduce the strength of the contact surface, but keep the flow field pressure high for a long time of more than 1 ms to give enough energy for ignition. From these results the following three conditions can be proposed for auto-ignition:

(1) After the pressure rises by the head shock and a little of pressure drops, the further pressure rise by the contact surface appears probably due to vortices which keep energy then temperature high in the flow field;

(2) Although the pressure rise by the contact surface is not strong, the probable vortices provide a longer pressure and them temperature conditions at the region between the head shock and the contact surface;

(3) The flow field pressure between the head shock and the contact surface becomes more than 1.5 MPa.

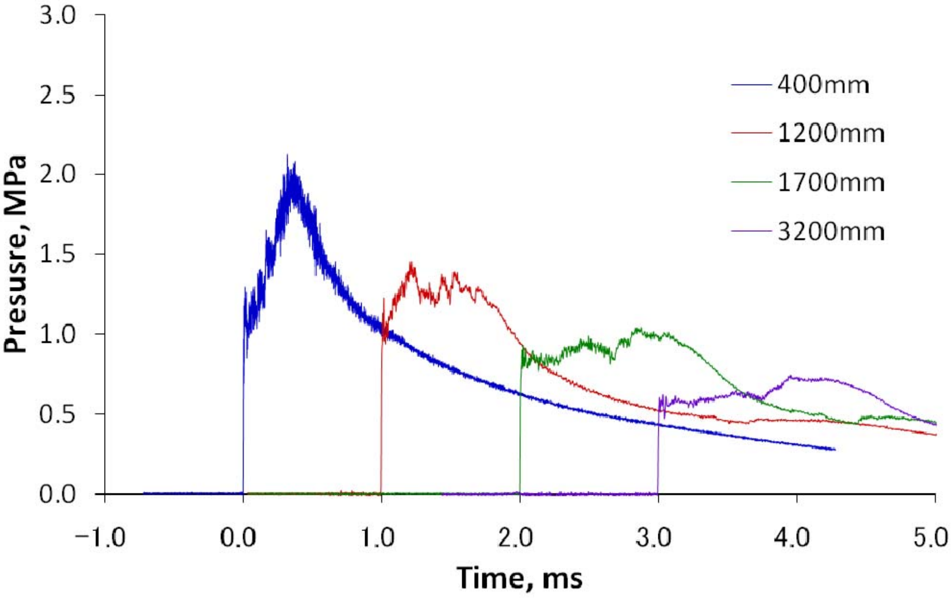

Fig. 2 Pressure histories of the pressure sensor P1 for four tube length cases

at the burst pressure of 4.8 MPa

3.3 Experiments of auto-ignition at the constant tube length when the diaphragm burst pressure is variable

When the shock tube length and the location of the pressure sensors are kept (Table 3), the maximum pressure becomes higher when the diaphragm burst pressure sets higher as expected. The further results are that the auto-ignition occurs at the contact surface pressure of higher than 1.5 MPa, which is obtained at the constant diaphragm burst pressure with variable tube length cases. The arrival time between the head shock and the contact surface is the shortest at the diaphragm burst pressure of 5.5 MPa. The cases of the diaphragm burst pressure higher and lower than 5.5 MPa give the longer arrival time between the head shock and the contact surface, where the diaphragm burst pressure of higher than 5.5 MPa gives the auto-ignition and that of lower than it gives the non-ignition situation. The reason for the phenomena is probably that the cases of the diaphragm burst pressure lower than 5.5 MPa simply do not have enough pressure for ignition, then the arrival time between the head shock and the contact surface becomes longer. On the other hand the cases of the diaphragm burst pressure higher than 5.5 MPa have a stronger head shock to provide bigger vortices and to slow down the arrival time for auto-ignition than the case of the diaphragm burst pressure of 5.5 MPa, which gives the right amount of pressure and shortest time for auto-ignition. The ignition and non-ignition situation may come from that the vortex effects are there or not and may be turbulent. These discussions basically come from the numerical results obtained by Yamada et al. [9] and not from the experimental observation.

Table 3 Conditions at the same tube length L of 300 mm

No	Tube length, mm	Diaphragm burst pressure, MPa	Thickness of diaphragm, μm	Mean shock speed, m/s	Auto-ignition
1	300	1.0	25	664	×
2	300	2.0	75	817	×
3	300	3.0	100	911	×
4	300	3.5	100	938	×
5	300	4.5	125	1063	×
6	300	5.5	150	1072	○
7	300	6.3	150	1157	○
8	300	7.5	188	1265	○

3.4 Reproducibility of the experiments

In order to check the reproducibility of the experiments, the experiments using the same conditions are performed 6 times as shown in Table 4. The experimental conditions for this check is the tube length of 650 mm and the burst pressure of 6.3 MPa and provide the mean shock speed between 1100 and 1200 m/s and auto-ignition for all cases. The pressure profiles of these six cases show almost same profiles. If there is some difference, the burst of diaphragm should not be different all the time.

Table 4 Reproducibility of the experiments using a condition of the tube length of 650 mm and the burst pressure of 6.3 MPa

No	Tube length, mm	Burst pressure, MPa	Thickness of diaphragm, μm	Mean shock speed, m/s	Auto-ignition
1	650	6.3	150	1192	○
2	650	6.3	150	1204	○
3	650	6.3	150	1132	○
4	650	6.3	150	1206	○
5	650	6.3	150	1157	○
6	650	6.3	150	1111	○

3.5 Relationship between shock wave speed and burst pressure

The relationship between the mean shock speed, c_s and the burst pressure is theoretically and in the first order calculated using the following equation from the shock wave relation [11] and is compared with the experimental results:

$$c_s = a_1 \left(\frac{\gamma - 1}{2\gamma} + \frac{\gamma + 1}{2\gamma} \frac{p_2}{p_1} \right)^{\frac{1}{2}}$$

where c_s is the head shock velocity, a_1 the speed of sound ahead of the head shock, p_1 the air pressure ahead of the head shock, p_2 the diaphragm burst pressure (p_b), and γ the specific heat ratio of air..

This equation comes from the shock wave relation between the inflow and out flow at the head shock. Hence the diaphragm burst pressure p_b does not match exactly with p_2 in the equation. When the tube length is short, the value of p_2 is close to p_b, but it becomes long p_2 is far from the value of p_b in many reason. Hence the equation may assure the head shock speed in a first order manner for the short tube case.

The experimental results using relatively the short tubes such as 300 mm and 650 mm give 100-200 m/s different from the theoretical results as shown in Fig. 3 and provide the similar results obtained by Mogi et al (2009, [7]) who used the results using the electric valve to spout high pressure hydrogen jet. In the cases of the tube length of 2.2-2.3 m, the shock wave speed becomes 300-400 m/s lower than the theoretical one due to many reasons such as the viscous effect with the tube wall and the vortices. Obviously the experiments with the short tube give the closer results to the theoretical ones as described above.

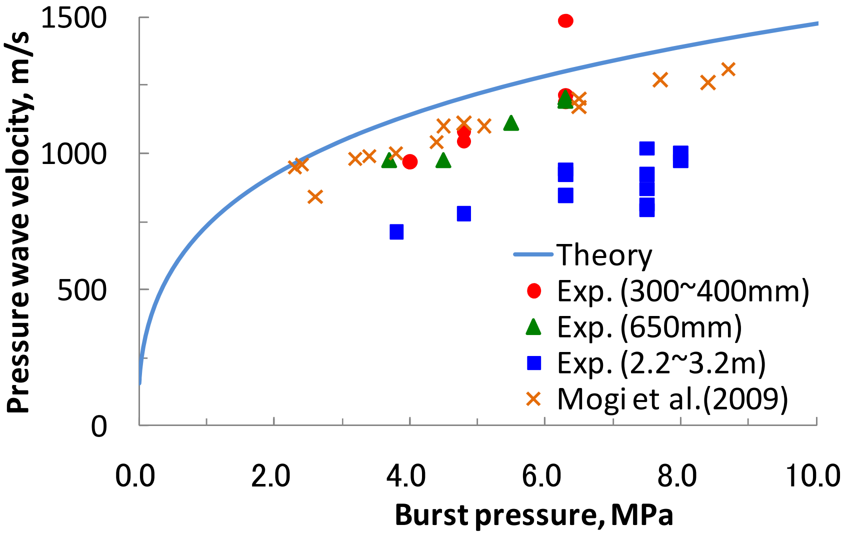

Fig. 3 The relation between the pressure wave velocity and the burst pressure and the comparison of them with the theoretical result at the various tube lengths

3.6 Map of auto-ignition limit

An auto-ignition map is created in the present experiment using the shock tube of various length and diaphragm burst pressure, which is shown in Fig. 4. The most important and interesting results the present study provides is that there is a minimum diaphragm burst pressure and a certain tube length for auto-ignition, which was not clearly obtained at the previous study by Mogi et al. [7].

Figure 4 shows the map of auto-ignition for the present experimental results and that of Mogi et al. [7]. The red solid circle plots are obtained using a plunger; the green ones are done by simply breaking diaphragm with a high pressure; the blue ones are obtained by Mogi et al. [7]; and the purple ones are by Pinto et al. [12]. The circles imply auto-ignition; the triangles are blow-off cases which are the case that the emission is recognized, but does not become a flame afterward; and the crosses are failed-ignition. The red plots in the present results show the straight at the same diaphragm burst pressure, which implies the experiments are quite accurate to keep the same diaphragm burst pressure and the use of plunger system works good for the present auto-ignition study.

It is understood from the present results that the minimum burst pressure exists at a longer tube length than 1 m in hydrogen jet coming out from tubes, but the past studies such as Mogi et al. [7] and Pinto et al. [12] did not show such minimum value since they used the tube length of less than 1 m. The apparent auto-ignition is not obtained at the cases of the tube length of more than 2 m. When the tube length is longer than the critical value; say like about 1.2 m in the present case, the loss of momentum and energy of the head shock and the contact surface together with the supposed vortices becomes more than ignition energy. If the diaphragm burst pressure is much higher, the critical tube length becomes longer. Since no tube length case (a jet coming out of a hole) does not give an auto-ignition, we do not get a finite diaphragm burst pressure at the tube length of zero in Fig.4. This result suggests that the longer tube is safer than the shorter one when the tube is set for the hydrogen tank.

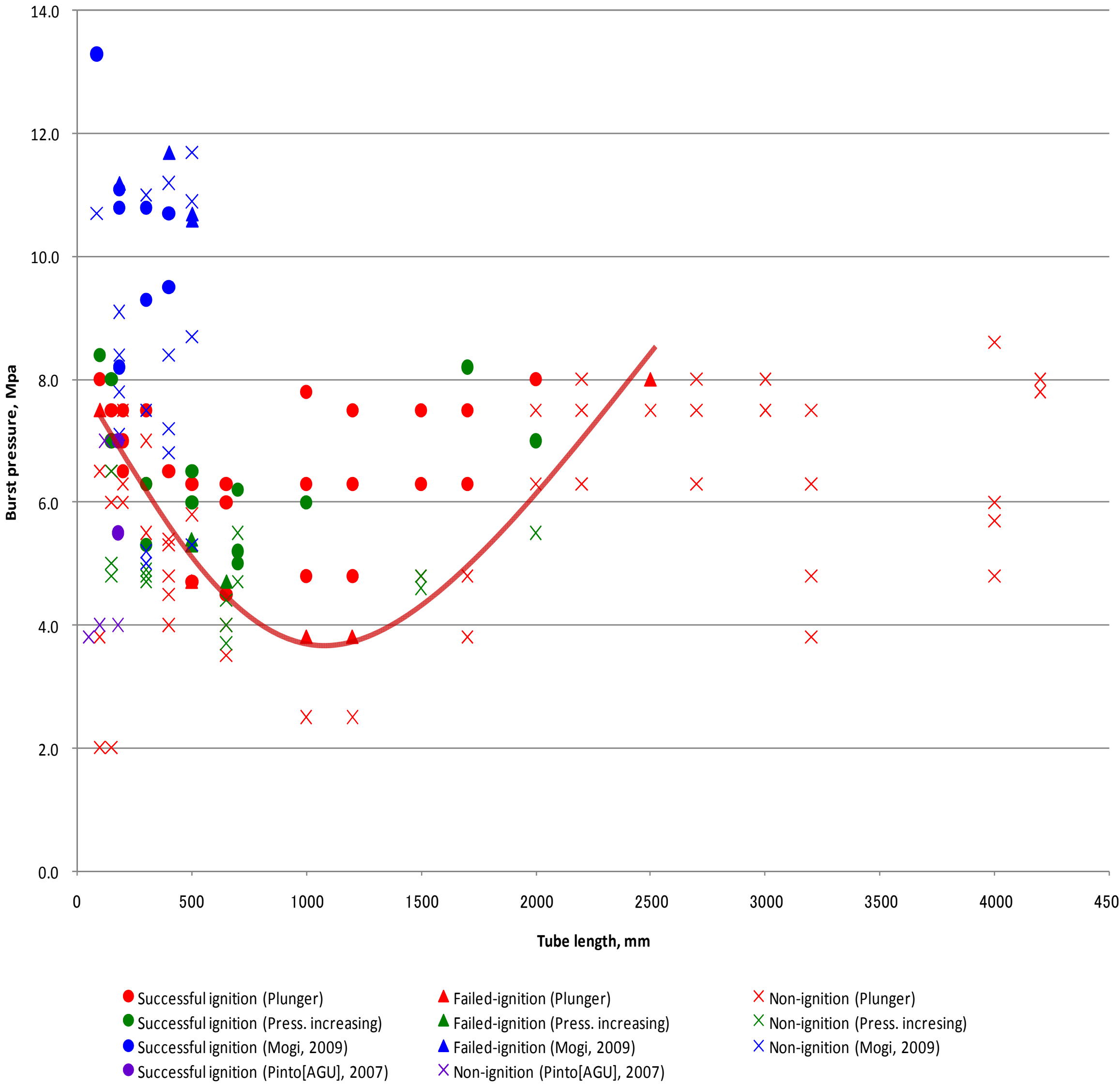

Fig. 4 Auto-ignition map described by the relation between tube length and burst pressure

3.7 Visualization of auto-ignition using a high-speed camera

A high speed camera is used to visualize whether hydrogen jet auto-ignites or not and to visualize the detailed flow feature of hydrogen jet near the tube exit. The measurement conditions and the performance of the NAC MEMRECAM GX-8 camera are described in Table 5. Figure 5 shows the sequential pictures of the hydrogen jet spouting from the tube exit at the burst pressure of 5.9 MPa and the tube length of 650 mm. The white part of the left end of the picture is the exit of the tube with the outer diameter of 20 mm (the tube inner diameter is 10 mm). The frame speed of 100000 frame/s is used to detect the Mach disc at the exit of the tube. The Mach disc starts appearing at the time of 30 μ s until about 100 μ s due to a supersonic expansion of the hydrogen jet, which disappears at about 110 μ s because hydrogen flow goes down. The white part in the pictures comes out clearly at the tube exit at the time of 30 μ s, which is probably a flame ignited in the tube and continues to be there until the time of 120 μ s. The auto-ignition also comes out behind (downstream of) the Mach disc due to high temperature. The shape of the flame behind the Mach disc is a half moon-like which is not seen for the result of the 2D numerical analysis, since the arc in 2D results becomes what we see in these pictures. Hence both the experimental results and the numerical ones agree each other. These pictures do not show the ignition near the exit, which was seen in 2D numerical results (Yamada et al. [9]), but such ignition was recognized in the other case.

Table 5 Conditions of high speed camera; NAC GX-8

Camera	NAC GX-8
Focal length, mm	105
F-number	2.8
Resolution	144x100
Sampling rate, fps	100000
Shutter speed, μs	9.2
Burst pressure, MPa	5.0
Tube length, mm	650
Thickness of diaphragm, μm	150
Mean shock speed, m/s	1031

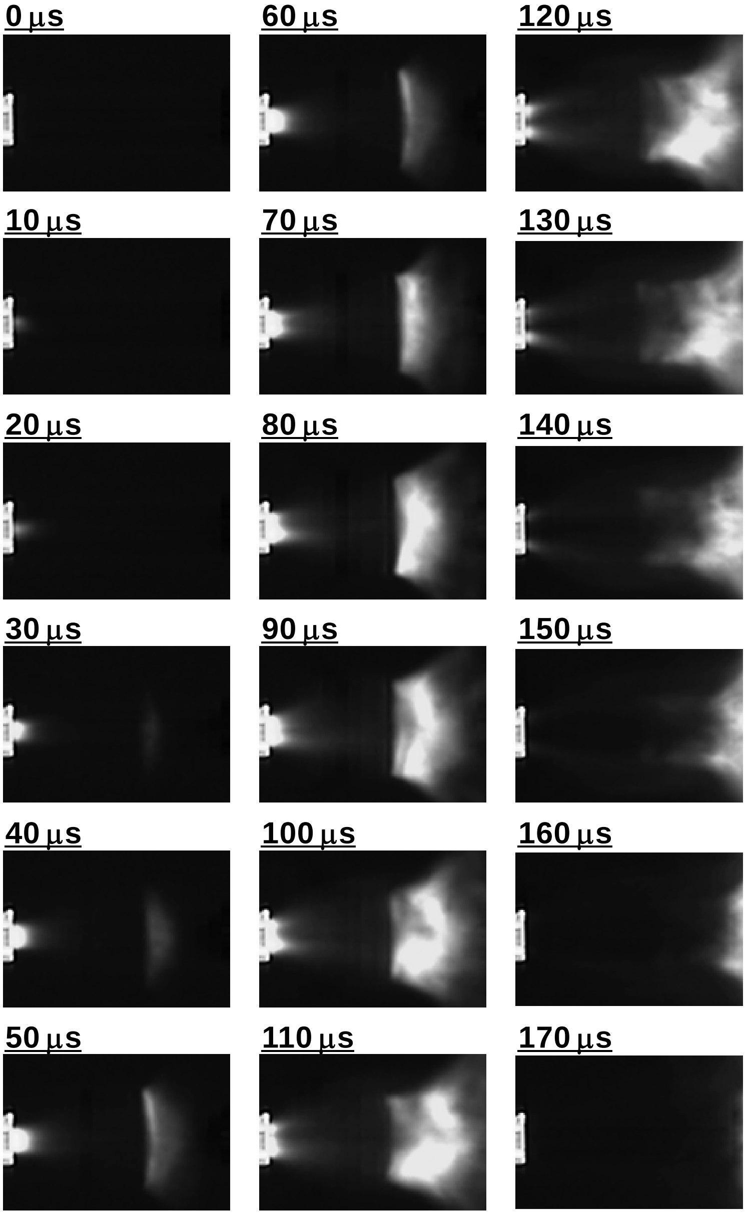

Fig. 5 High speed sequential pictures of the hydrogen jet, flame generation, and its propagation at tube exit of the tube (L = 650 mm, Pb = 5.0 MPa)

4.0 CONCLUSIONS

The various lengths of shock tube are used to study auto-ignition of hydrogen jet spouting out of the tube at several different diaphragm burst pressures.

The most important result in this experiment is that the auto-ignition occurs at the higher diaphragm burst pressure than the critical and minimum burst pressure which in the present case is about 3.8 MPa and the tube length of about 1100 mm. Below this critical diaphragm burst pressure and with the longer or shorter than the tube length of about 1100 mm, no auto-ignition occurs.

Then the flame shape obtained in the 2D numerical results are confirmed in the high speed pictures using the high speed camera with 100000 frames/s.

ACKNOWLEDGMENTS

Dr. Hiromi Shiina and Mr. Teruo Ebuchi of National Institute of Advanced Industrial Science and Technology, are appreciated for their experimental suggestion and support for this research.

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