# Self-ignition and Flame Propagation of Pressurized Hydrogen by Sudden Release through a Tube

Hyoung Jin Lee<sup>1</sup>, Sei Hwan Kim<sup>2</sup>, Yeong Ryeon Kim<sup>2</sup>, In-Seuck Jeung<sup>2</sup> <sup>1</sup> LIG Nex1 Co., Ltd., Daejeon, Republic of Korea <sup>2</sup> School of Mechanical and Aerospace Engineering, Seoul National University, Seoul, Republic of Korea

# **1** Introduction

Hydrogen is regarded as a one of future fuels which will take place of oil fuels. As the popularity on the hydrogen is increased, the crucial safety problems concerned with utilization of hydrogen are storing and transportation. Due to low energy density in gas phase, hydrogen needs to be stored under very high pressure or in liquid state. Because the latter storage system is much more complex and expensive, high-pressurized hydrogen has been used. For example, hydrogen fuel-cell vehicles may possess storage pressures as high as 70 MPa.

However, high pressure hydrogen installation is endangered by a risk of sudden discharge of hydrogen leading to ignition and severe accident. It has been reported that a leak of such high-pressurized hydrogen into the air can lead to self-ignition and explosions without any apparent ignition sources. The incidents have been reported by Astbury and Hawksworth [1]. For this reason, many studies have been conducted to understand this phenomenon and obtain the criterion to be occurred self-ignition.

The first investigations concerned with a problem of high pressure hydrogen outflow ignition were carried out nearly 40 years ago by Wolański and Wójcicki [2]. Following the Wolanski and Wojcicki's experiment in which a shock tube was used, diffusion ignition mechanism has been one of the most primary concerns relating to this issue. Dryer et al. [3] discovered that spontaneous ignition requires both a sufficiently high bursting pressure and a downstream geometry for fast mixing of the expanding hydrogen and shocked air. Additionally, they postulated that turbulent mixing and the heating of the mixture by the multi-dimensional shock-boundary and shock-shock interactions might play a significant role in the generation of an ignition. Recently, this postulation has been proven by numerical simulation of Lee and Jeung [4] and experimental observations of Lee et al. [5]. They showed that two reaction regions inside the tube, where one reaction region is in the boundary layer and the other is in core region of the tube due to shock-shock interactions, can be generated and selfignition can occur when two reaction regions are completely merged. Especially, using direct images, Lee et al. showed that there can be failed-ignition cases which the flame cannot sustain when it goes out to the open air, although the spontaneous ignition occurred. In addition to these efforts, several experiments have been conducted to identify the limiting conditions of self-ignition. Mogi et al. [6,7] conducted several experiments varying the length of tubes from 3 mm to 500 mm and bursting pressures up to approximately 20 MPa. Lee et al. conducted similar experiments using commercial rupture disk system for practical usage. They confirmed that the self-ignition can occur at the bursting pressure of 23.5 MPa with short tube of 50 mm [5]. The previous results have shown that the hydrogen

### H.J. Lee et al.

jet had an increasing tendency to ignite as the downstream tube increased in length. In spite of these valuable results which the mechanism of self-ignition can be uncovered, the investigations outside the tube have been rarely reported, such as the flame propagation, flame stabilization and extinction. In this study, the experimental apparatus comprised of commercial rupture disks to simulate actual circumstances and tubes with an inner diameter of  $5 \sim 10.9$  mm and lengths ranging from 35 mm to 600 mm. This paper presents values of the minimum bursting pressures for self-ignition with the tube lengths and the effect of tube diameter on the self-ignition. Additionally, using the direct and shadow images outside the tubes, we investigated the flame propagation, flame stabilization and extinction based on the experimental data.

# 2 Experimental setup

A schematic of the experimental apparatus is shown in Fig. 1. The main apparatus consists of hydrogen storage tanks, a disk holder, a rupture disk and a downstream tube. Figure 2 shows the combination of a disk holder, a rupture disk and a downstream tube. The commercial rupture disk for 1/2 inch (Fine disk, Korea) which is made by SUS is inserted inside the disk holder and fixed and sealed by a disk ring which has tapered shape. The diameters of a disk ring and an extension tube are the same as 10.9 mm. The upstream and downstream diameters of the rupture disk are the same. The experiments were conducted varying the extension tube length from 35 to 600 mm.



Figure 1. Schematic of the experimental apparatus.



Figure 2. Combination of a disk holder, a rupture disk and a downstream tube

The test procedures in the experiment are as follows: at first, air in the two storage tanks is removed using a vacuum pump and then hydrogen is pressurized at the first storage tank to wanted pressure (maximum about 1,000 atm) by using an air driven gas booster (Haskel, AG-152). The volume of first storage tank is 2 liter and that of the second tank is 0.5 liter. After that, hydrogen feeds to the second storage tank from the first storage tank through a pipe until a rupture disk bursts, thereby resulting in the sudden release of hydrogen into the atmosphere through a downstream extension tube. The burst

#### H.J. Lee et al.

#### Self-ignition and flame propagation of pressurized hydrogen

pressure can be known properly because only peak value is set to be recorded. Figure 3 shows the picture of experimental apparatus.



Figure 3. Experimental apparatus

In order to investigate the phenomena occurring inside the tube, two types of measurement device are installed. In order to measure the pressure and propagation of a pressure wave, several piezoelectric pressure transducers (PCB Piezotronics, 113A22) are installed at the tube wall. The first pressure transducer is located at 35 mm from the rupture disk and sequence transducers are installed at every 50 mm as shown in Fig. 4. Figure 4 is an example of extention tube of 300 mm. Furthermore, in order to observe the flame propagation, the photodiodes (Panasonic, PNZ 300F) as a light detector are installed at the opposite side wall from the PCB sensors, as shown in Fig. 4 The bust pressure of a rupture disk is measured using a pressure transducer (Sensys, PMHA 1000). After the rupture disk bursts, the shock wave formed inside the tube and the voltage signal is outputted when the shock wave pass through the installed pressure transducers. The rising signal from the first transducer is used as a trigger signal of the measuring system such as data acquisition system and high speed camera. The output signals from the pressure sensors and photodiodes are recorded by using a data acquisition system. The ignition and flame propagation phenomena outside the tube are recorded by using two high-speed digital video cameras (Phantom, v710). One is for the schlieren images and the other is for dirct images as shown in Fig. 1. The high speed camera records at every 10 µsec and frames of maximum 55,000 can be recorded before and after the trigger signal.



Figure 4. Drawings of downstream tubes and measurement positions

# **3** Results and Discussion

Figure 5 shows the ignition map as a function between burst pressure of the rupture disk and the extension tube length for three cases such as 1) successful ignition, 2) failed ignition, and 3) no ignition of the jet flow. Failed ignition signifies that the flame blows out though the self-ignition

occurs. And Figure 5 also shows the illuminance data inside the extension tube when the burst pressure is 23.5 MPa and tube length is 200 mm with internal diameter of 10 mm.



Figure 5. Burst pressure for self-ignition(left) and Illuminance with time inside extension tube(right)

In the Fig. 5, when burst pressure increased, the possibility of self-ignition also increases even though the extension pipe is short. In case of previous experiments, there is no observation of self-ignition when the tube length (internal diameter of 10 mm) is about 50 mm. In this experiments, self-ignition is confirmed at the when the tube length (internal diameter of 10.9 mm) is 50 mm at the burst pressure of 235 atm. It appears that it is very difficult for a high-pressure hydrogen jet to get self-ignited when the pipe is open to the atmosphere. However, there are no experimental results above the burst pressure of 200 atm. Therefore, it is possible to self-ignite at higher burst pressure than 200 atm even though the tube length is short. The typical light emissions in the measurement tube are shown in right image of Fig. 5 when the hydrogen jet is ignited successfully. Time zero indicates the time when the first pressure transducer detects an increase pressure profile inside the tube. The emission from the flame increases as the flame approaches the pipe outlet. The presence of the flame inside the tube is confirmed from the emissions.

To study the flame propagation and ignition at the tube outlet, a high-speed camera is used to obtain the visual data. Fig. 6 shows the direct and shadow images outside a tube with a tube length and the burst pressure is the same of 23.5 MPa. The tube length of left image is 100 mm and corresponds to the failed ignition. The right is the case of self-ignition and tube length is 300 mm. The images are obtained from a side view by using a color high-speed camera. The interval time of the frames is 8.43  $\mu$ sec and the exposure time is 7.71  $\mu$ sec. The trigger time is when the output signal of the first pressure transducer begins to increase.

For the self-ignition case, the flame produced by self-ignition in the tube is coming out of the tube outlet, after which it propagates downstream. And then, another flame like a flat flame is observed in front of the heading flame. This new flame is larger than the tube diameter. This flame gradually grows and separates into two flames. Although one flame collapses, the other flame yields due to the ignition and the hydrogen jet flow is stabilized at the tube outlet. It is confirmed that the flame at the tube outlet is not a lifted one, and self-ignition is initiated at the tube edge. When the high-pressure hydrogen is spouting from the tube into still air, a semi-spherical shock wave is produced in the air. Then, the shock wave heats the air to a high temperature and induces the self-ignition of hydrogen at the length of the tube for the mixing of hydrogen and air is sufficiently necessary so that the self-ignition occurs. Thus, tube diameter affects the phenomenon of self-ignition. As a diameter of tube decreases the multi-dimensional shock interactions can take place inside a tube. This leads to the self-ignition at shorter tube length when the burst pressure is the same.



Figure 6. Direct image of flame propagation and Shadow graph image for failed-ignition(a) and self-ignition(b)

### 4 Summary

In this study, we investigated in detail the characteristics of self-ignition of high-pressure hydrogen jet during its sudden release by changing the tube length together with the flame propagation at the tube outlet. For the pressure range up to 20 MPa, it was very difficult for the high pressure hydrogen jet to get self-ignited when the downstream tube length is short. However, the possibility of ignition in the tube increases above about 10 MPa and self-ignition occurs with tube which has longer than 100 mm. From the experiment using the measurement inside the tube, the presence of a flame in the tube is confirmed, and when burst pressure increased, the flame is detected at a position near the rupture disk. Furthermore, the emission from the flame becomes strong as the flame approaches the tube outlet. At the tube outlet, the flame is not lifted and the self-ignition is initiated at the outer edge of the jet. On the basis of the present experimental results and discussions, it is suggested that there is a possibility of ignition in the tube when the high-pressure hydrogen gas is momentarily discharged through the tube. These results suggest that it is necessary to obtain the data of self-ignition for higher bursting pressures when working with shorter tubes. For tubes of insufficient length, the flame was observed only in the boundary layer, and subsequently weakened and extinguished after exiting the tube. A complete flame across the entire cross-section of the tube is essential for the initiation of self-ignition because it has the potential to sustain a diffusion flame after exiting the tube into the air. Furthermore, in order to establish a complete flame, sufficient tube length is necessary so that the mixing region is generated by multi-dimensional shock-shock interactions. For this reason, the possibility of selfignition increase as the tube diameter decreases. When self-ignition occurs successfully, the flame is not lifted at the tube outlet and self-ignition is initiated at the outer edge of the jet.

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