

EXPERIMENTAL STUDY ON THE SELF-IGNITION OF

PRESSURIZED HYDROGEN RELEASE INTO THREE-WAY TUBES

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ABSTRACT

To explore the effect of bifurcation structures on the spontaneous ignition and shock wave result from the sudden release of pressurized hydrogen. Three-way tubes with different bifurcation angles (90°, 120°, 150°) were used in the experiments. They are two Y-shape tubes and one T-shape tube. The photoelectric and pressure signals in the tube were recorded by the sensor. The results show that the reflected shock wave will be formed at the bifurcation. In addition, the intensity and velocity of the leading shock wave will attenuate sharply when it passes through the bifurcation. The smaller bifurcation angle of tube, the smaller overpressure decay rate of shock wave at bifurcation position. The smaller the bifurcation angle of tubes, the weaker the reflected shock wave transmitted downstream, and the greater attenuation of shock wave intensity. Experimental results have reference value for the safety of hydrogen storage at high-pressure, and are helpful to understand the influence of different tube structures on spontaneous ignition when hydrogen is transported at high pressure.

Keywords: Hydrogen safety, Pressurized hydrogen, Self-ignition, Bifurcation structures, Shock wave

INTRODUCTION

With the advent of the information age, more and more countries are gradually abandoning fossil energy, which causes environmental pollution, and are seeking new, cleaner and more efficient energy sources instead. Hydrogen regarded as an emerging energy source suitable for replacing fossil fuels in the 21st century due to its high combustion calorific value, non-polluting combustion products and its renewable character. However, the storage of hydrogen is a major challenge in today's energy applications. Nowadays, The more mature and commonly used method of hydrogen storage in the world is high pressure hydrogen storage, but high pressure hydrogen storage still has many dangers. Because of the rapid spread of high-pressure hydrogen and the wide combustion limits (4%-75%). These characteristics make the transport and storage of hydrogen extremely vulnerable to accidents such as fires and explosions, which can cause great damage to human life and property.

Many previous studies[1-17] have confirmed sudden release of high-pressure hydrogen into a tube even in the absence of an external ignition source can cause self-ignition. A variety of ignition theories have been proposed in response. The "diffusion ignition theory", proposed by Wolanski et al.[2] in 1972, is currently the most common hypothetical ignition theory. They created a simple model of diffusion ignition by building an experimental setup for hydrogen diffusion to oxygen. Dryer et al.[3] identified the use of downstream release tubes as a necessary condition for hydrogen self-ignition. Because the release tube provides sufficient space for stable leading shock wave formation and shock wave heating of the hydrogen-air gas mixture. Mogi[4,5] and Lee et al. [6] both found through experimental studies that high-pressure hydrogen is difficult to self-ignite in short

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downstream tubes by pressurising the storage tank, which in turn initiates the bursting of the bursting disk and the hydrogen gas enters the short tube. Golub et al. [7,8] reveal the internal mechanism of self-ignition caused by the bursting of high-pressure hydrogen gas through a burst disk and its sudden release into the tube through both experimental and numerical analysis. Xu et al. [9,10] found by simulating the release of high pressure hydrogen into downstream tubes of different shapes (partial constriction, partial expansion, sudden constriction and sudden expansion) that changes in shape of tubes or promote self-ignition of hydrogen in tubes. Gong et al. [11] investigated the effect of different angles of tube on ignition of high-pressure hydrogen releases by changing the angle of the downstream tube. They found that smaller angle between tube and direction of hydrogen jet, easier it is to create higher intensity shock waves. Morii et al. [12] found through numerical simulations that adding an obstacle to a downstream tube promotes hydrogen self-ignition. Many previous studies found that sudden release of high pressure hydrogen from a tube to atmospheric air creates leading shock wave at front of hydrogen jet [13-16]. Grune [15] and Duan et al. [16] found that multidimensional reflected shocks formed between leading shock and hydrogen-air contact surfaces in both their experiments.

In summary, it is found that experimental or numerical simulations at this time focus on straight or other shapes of two-way tubes. However, in actual hydrogen applications, in addition to straight tubes, Y-shape tubes with different angles or other irregularly shaped three-way tubes are mostly used. Therefore, in this experiment, high-pressure hydrogen gas releases into three-way tubes at different angles via natural membrane rupture. Investigation of high-pressure hydrogen self-ignition in tubes by changing initial burst pressure. To investigate the relationship between initial burst pressure and leading and reflecting shock wave intensities, and to discuss mechanisms of self-ignition in tubes.

EXPERIMENT SETUP AND METHODS

Experiment set up

Fig.1 shows the experimental setup for high-pressure hydrogen release to downstream tubes via rupture of the bursting disk. The experimental setup consists of upstream gas supply system (nitrogen gas cylinder, hydrogen gas cylinder, 1- pressure reducing valve, 2- high-pressure pneumatic valve, 3- controller), high-pressure hydrogen storage system (inlet tube, relief valve, 4- tank pressure gauge, 5- high-pressure storage tank, 6- vacuum pump), downstream relief system (burst disk, downstream tube, 7- burst disk holder), data acquisition system (photoelectric sensors L1-L4 and pressure sensors P1-P4, signal amplifier, 8- data acquisition equipment, 9- high-speed camera, 10- computer). The storage tanks, tubes and tube connectors are made of 316L stainless steel. The whole setup is designed for maximum pressure of 30 MPa. Hydrogen of 99.999% purity used as experimental gas. Nitrogen of 99.999% purity used for pre-testing experimental systems, checking gas tightness of devices and post-test purging of exhaust gases.

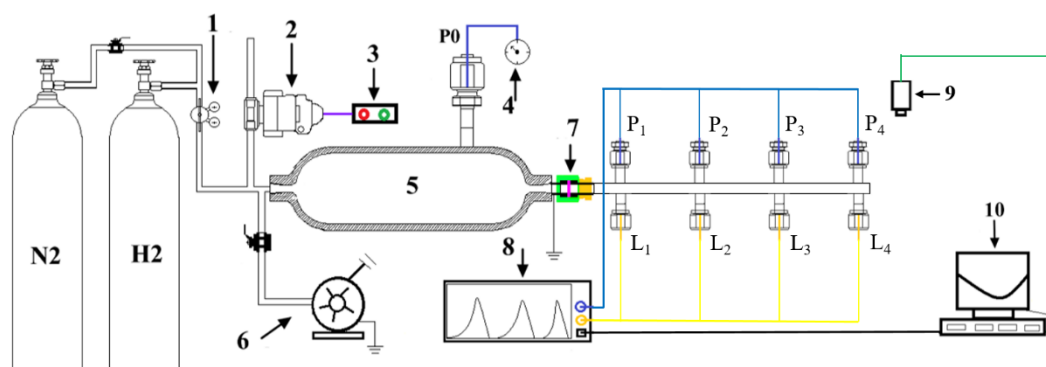
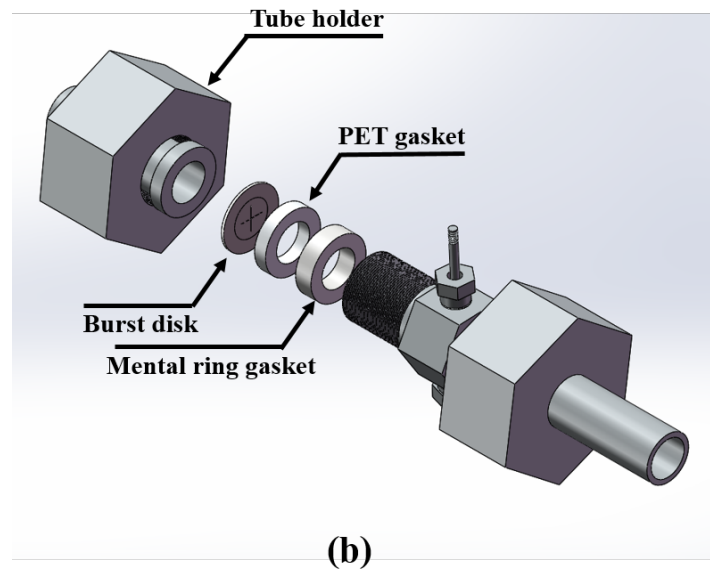
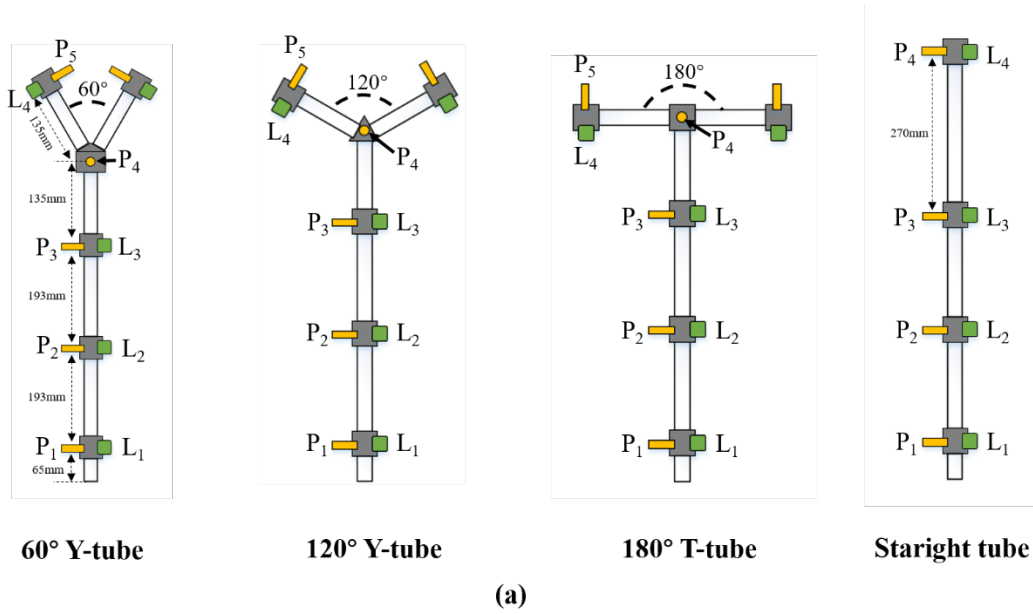


Fig.1. Schematic of experimental apparatus: 1 Pressure reducing valve, 2 High-pressure pneumatic valve, 3 Controller, 4 Tank pressure gauge, 5 High-pressure storage tank, 6 Vacuum pump, 7 Burst disk holder, 8 Data acquisition equipment, 9 High-speed Camera, 10 Computer

Fig.2(a) shows a diagram of downstream tube connections. Tube diameter 15mm. The tube consists of two 193mm long straight sections and a 135mm three-way tube. The distance from the burst disk to the first sensor (P1 and L1) is 65 mm. The outlet part of the tube is two symmetrical openings in three-way tubes (i.e. tube nozzles). The distance from the burst disk to tube nozzle, i.e. the total length of three-way tube, shown in Figure 2(a). The burst disk installation is shown in Fig.2 (b) and (c). At the three-way bifurcation position, a single pressure sensor P4 is installed to record the pressure changes in bifurcated sections of tube. Pressure sensors (PCB-113B22, USA) P1, P2, P3, P5 and photoelectric sensors (Thorlabs,FDS-010) L1-L4 are symmetrically distributed on downstream tube walls. Four different angles of downstream tubes are set up for this experiment: 60°, 120°, 180° and straight tube. The straight tube is made up of two 193mm sections with a 270mm section and only four symmetrical sets of sensors.



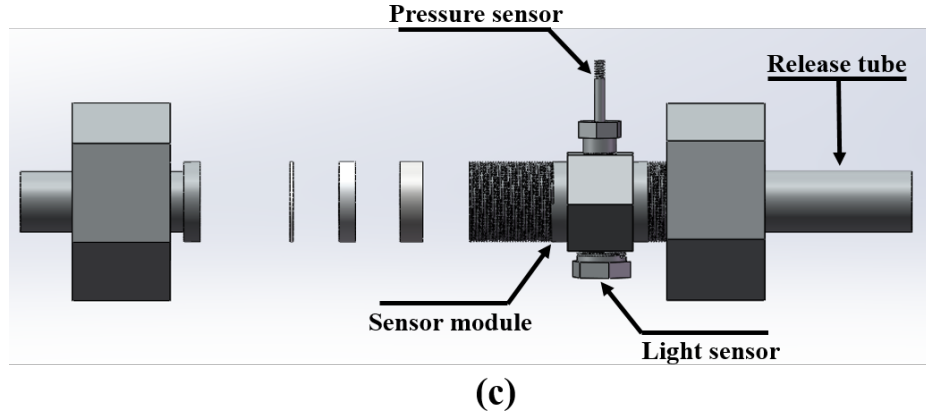


Fig.2. (a) Downstream tube connection (b) Overall view of bursting disk installation structure
(c) Front view of bursting disk installation structure

Experiment method

First, experiment by closing the 2- high-pressure pneumatic valve and installing the burst disk. Then, 6-vacuum pump is used to evacuate 5- the high pressure tank and tube upstream of the disk. Start 8- Data acquisition equipment and then open the hydrogen cylinder valve. After that, open Valve 1 and inject hydrogen gas until the burst disk ruptures. Recording the initial burst pressure P_0 when the burst disk ruptures. Meanwhile close valve 1 and hydrogen cylinder valve immediately, then open valve 1 and open nitrogen cylinder valve for purging. When final purging finished, close the nitrogen valve, close valve 1, save the data and prepare for the next set of experiments. The vacuum level at the end of evacuation during the experiment is approximately -1.0 atm. The inlet gas flow rate to the main vessel in these experiments is approximately $0.6\text{m}^3/\text{min}$.

Table.1 shows the tube and initial burst pressure for this experiment, with P_0 ranging from 3 to 8 mpa.

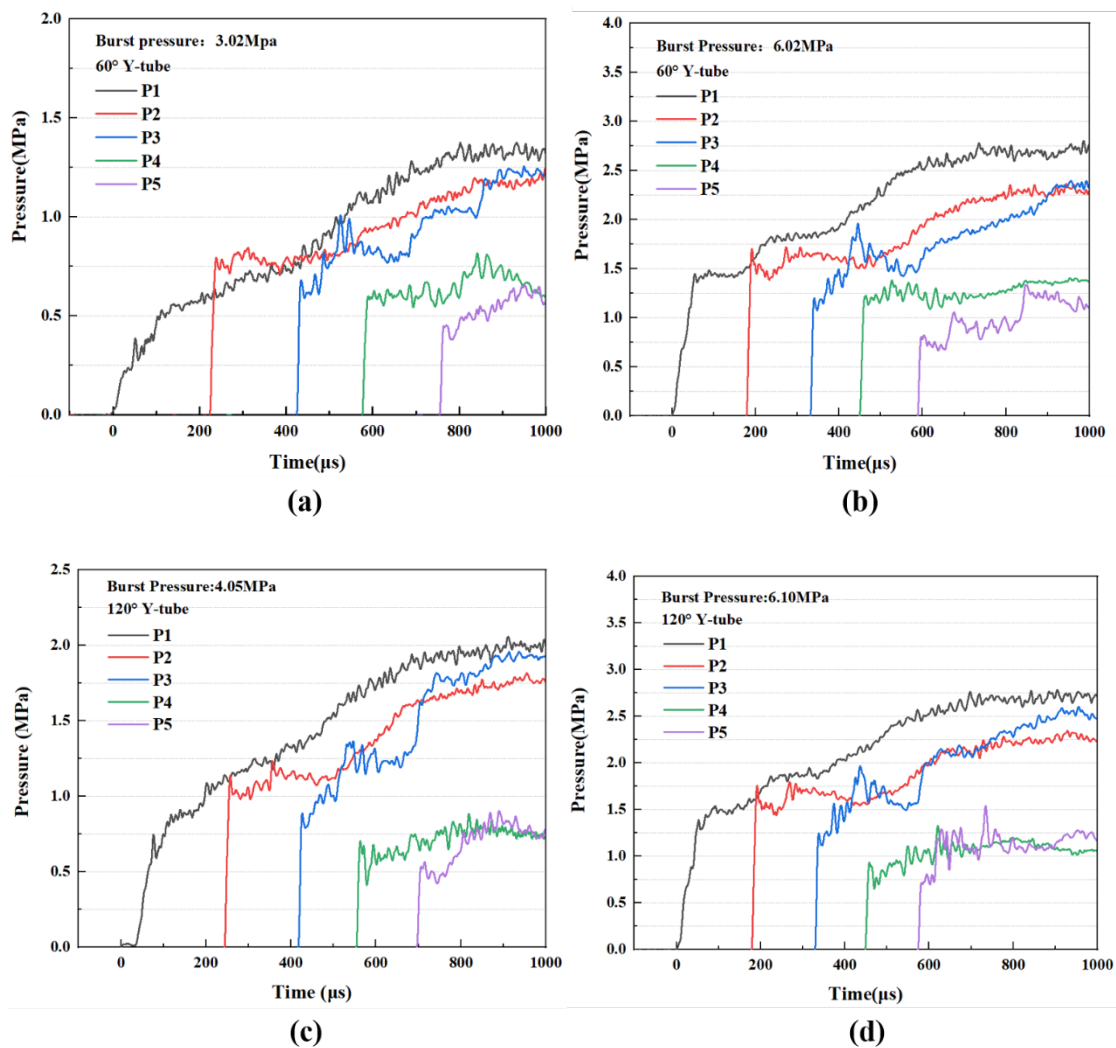
Table.1. Tube type and initial burst pressure P_0

Tube type	Burst Pressure(MPa)								
60°Y-tube	3.02	3.98	4.05	4.97	6.02	7.09	7.99	8.10	-
120°Y-tube	2.98	3.12	3.88	4.05	5.01	6.10	7.09	8.05	8.12
180°T-tube	2.98	4.01	4.15	5.09	6.02	6.11	6.95	7.15	8.03
Staright tube	3.03	3.89	5.04	6.07	-	-	-	-	-

RESULTS AND DISCUSSION

Fig.3 shows the pressure data in tubes for high-pressure hydrogen gas released into downstream tubes at different bifurcation angles. Define the time for signal to be received by P1 pressure sensor as $t_0 = 0\mu\text{s}$. In Fig.3(a) and (b) P1 at burst pressure of $P_0 = 3.02\text{ MPa}$ has more gentle rising trend than P1 at $P_0 = 6.02\text{ MPa}$. The reason for that is because leading shock waves are strong compressional waves and can be thought of a series of weak compressional waves superimposed[17]. Therefore the

development of leading shock is a gradual process. In contrast, when P_0 is greater, the leading shock wave is formed more quickly. In Fig.3, the pressure decay in tube is different for different bifurcation angles. The pressure decay at the bifurcation of 120° Y-tube is greater than in 60° Y-tube. This is because when the tube bifurcation angle is smaller, the angle α between tube wall and gas flow direction becomes smaller and the contact surface between high-pressure gas and tube wall becomes smaller. However, as both the 60° and 120° tube walls are not perpendicular to the gas flow direction, the reflected shock waves formed at tube bifurcation locations (P4) that transmit in the opposite direction to hydrogen jets are slight. In this case the gas is suppressed by expansion waves more than the facilitation of multiple wave structures such as reflected shock waves. Therefore the greater pressure decay of 120° Y-tube at bifurcation site.



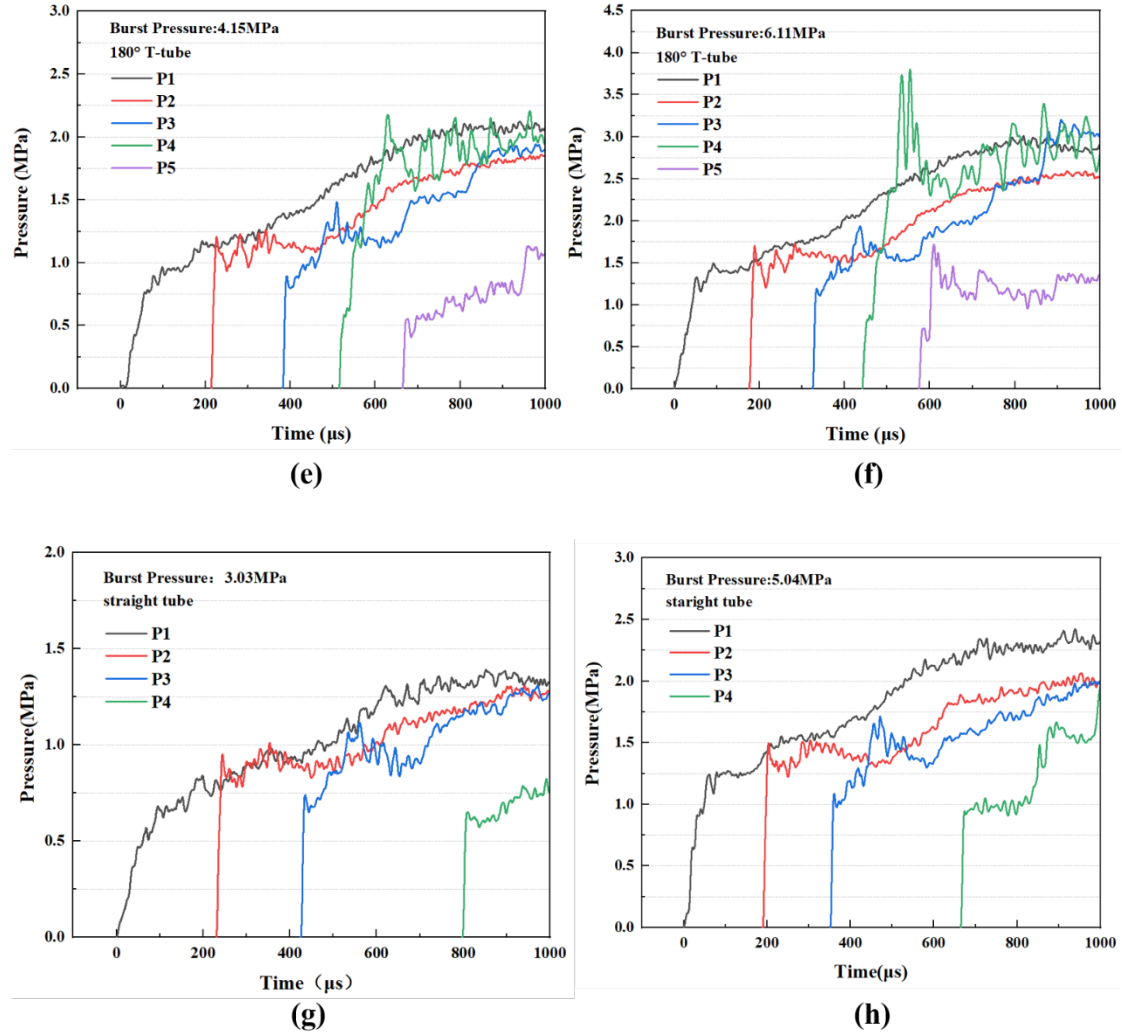


Fig.3. Pressure in tube at different tube types as a function of time

The pressure decay in 120° Y-tube after the wave passes through the bifurcation is less than in 60° Y-tube. The reason for this is that as angle α between tube and gas flow direction increases, the area where leading shock waves hit tube walls becomes larger and most of them form reflected shock waves transmitted downstream, at this time leading shock waves, reflected shock waves and other complex multi-dimensional shock waves are superimposed in the radial direction, making the pressure data of 120° Y-tube less decayed after bifurcation position. Table 2 shows the overpressure data of different tube types. The formula for calculating the overpressure decay rate is: $\frac{P_3 - P_4}{P_3} \times 100\%$. In Table 2, the decay rate of overpressure of shock waves becomes larger as the angle of tube increases. However, in Fig.3 there are several sudden rises in P4 of 180° T-tube and later P4 is larger than P3. Because the wall of 180° T-tube is perpendicular to the direction of gas flow. When leading shock waves are spreading to the bifurcation position. Positive shock wave hits the tube wall directly, creating a strong reflected shock wave. Moreover the velocity at this point P4 is reduced to 0 by hitting. The superimposed effect of shock waves causes P4 pressure to increase to very high levels. However, at this point the overpressure of P4 is only suppressed by expansion waves and thus reduced.

Table.2.Shock wave overpressure in different tube types

Tube type	Burst pressure(MPa)	P ₃ (MPa)	P ₄ (MPa)	Overpressure decay rate(%)
60° Y-tube	3.02	0.68	0.60	11.8
	3.98	0.81	0.79	2.5
	4.97	1.13	1.12	0.88
	6.02	1.20	1.22	-1.7
	7.09	1.37	1.54	-12.4
120° Y-tube	2.98	0.19	0.15	21.1
	4.05	0.89	0.70	21.3
	5.01	1.13	0.84	25.7
	6.10	1.24	0.93	25.0
	7.09	1.35	1.03	23.7
180° T-tube	3.02	0.70	0.45	35.7
	4.15	0.89	0.59	33.7
	5.09	1.08	0.75	30.6
	6.11	1.19	0.82	31.1
	6.95	1.26	0.89	29.4

Fig.4 shows the change in average spread velocity of leading shock waves as high-pressure hydrogen gas is released through different tube types. The average velocity of shock wave spread is calculated from the ratio of the distance between two adjacent pressure sensors and difference in arrival time of leading shock waves. In Fig.3 and Fig.4, as initial burst pressure increases, the intensity of shock waves detected by pressure sensors increases and the average velocity of spread of leading shock waves increases. In Fig.4, the average velocity drop of 60° Y-tube leading waves is greater than 120° Y-tube and 180° T-tube as leading waves transit tube bifurcation locations. The reason for this is that when the supersonic airflow is disturbed by tube wall, expansion wave is formed, resulting in reduction in pressure but corresponding acceleration in airflow velocity. So the average velocity of spread of leading shock waves between 120° Y-tube P4 and P5 rises.

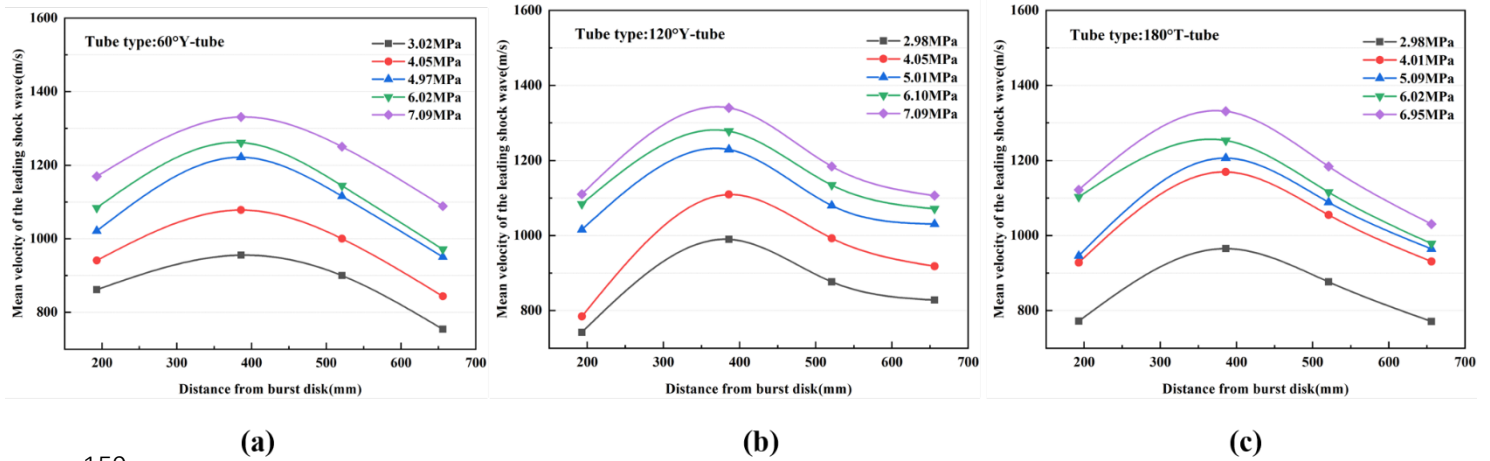


Fig.4. Average velocity of spread of leading shock wave in different tube types

Fig.5 shows the self-ignition of high-pressure hydrogen gas inside and outside tube after the release of the tube at different bifurcation angles. In Fig.5, the minimum initial burst pressure P_0 to cause spontaneous ignition in 60° Y-tube, 120° Y-tube and 180° T-tube is 7.09 MPa, 6.10 MPa and 5.09 MPa respectively. With the increase of bifurcation angle, the initial critical pressure that can cause spontaneous ignition of high-pressure hydrogen is lower and spontaneous ignition is more likely to occur. This is because when the initial burst pressure is small, the intensity of shock waves is not sufficient to cause spontaneous ignition of high-pressure hydrogen. The higher initial burst pressure of 60° Y-tube that causes spontaneous ignition of high-pressure hydrogen is due to the fact that expansion waves have a greater suppressive effect on the onset of spontaneous ignition than the promotion of multi-dimensional shock wave structures such as reflected shock waves.

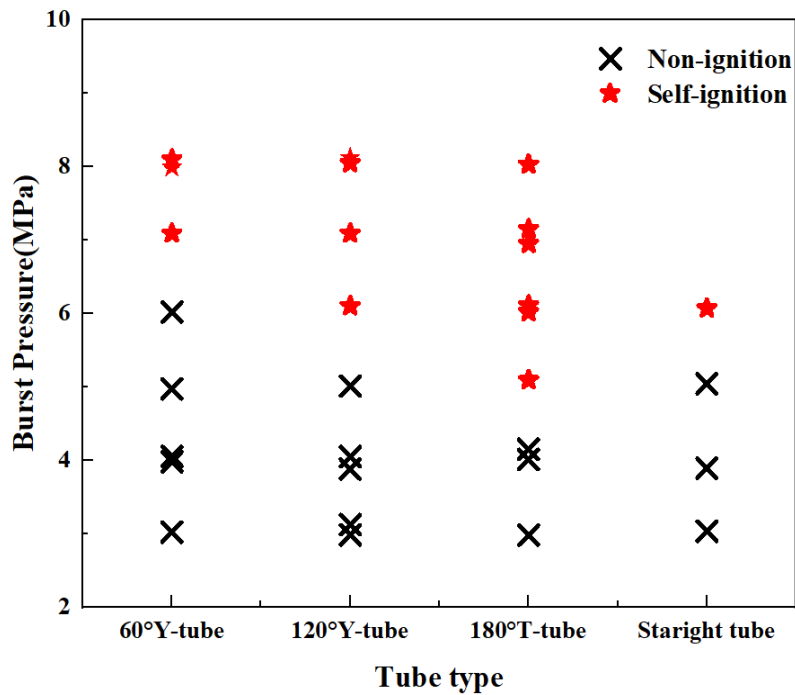


Fig.5. Self-ignition of different tube types

CONCLUSIONS

In this paper, a high-pressure hydrogen burst disk is used to naturally rupture the membrane and release it into the tube at different bifurcation angles. Analysis of pressure changes in tube shock and spontaneous ignition of high-pressure hydrogen by changing the initial burst pressure. Investigating the effect of bifurcation angle on high-pressure hydrogen self-ignition. Concluded the following 3 conclusions:

1) At different bifurcation angles of the tube, the lower initial burst pressure, the lower risk of self-ignition of high-pressure hydrogen. The lower bifurcation angle of tube, the higher critical pressure that can cause self-ignition, and the more difficult it is for self-ignition to occur.

2) In different bifurcation angles of tubes, the greater initial burst pressure, the greater intensity of leading shock waves and the faster leading shock wave formation. As high-pressure hydrogen gas transits the bifurcation location of tube, the intensity of leading shock waves is significantly reduced. The smaller bifurcation angle of tube, the smaller overpressure decay rate of shock wave at bifurcation position. After the high-pressure hydrogen passes through the tube bifurcation position, the smaller of tube bifurcation angle, the weaker reflected shock wave spreads downstream and the greater decay of shock wave intensity.

3) At different bifurcation angles, the average speed of leading shock wave spread is faster when initial burst pressure is higher. The smaller bifurcation angle of tubes, the faster average velocity of leading shock waves drops after the bifurcation position.

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