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Ignition of Hydrogen Suddenly Leaked from a High Pressure Tank

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[Abstract] When high pressure hydrogen jet is released to atmosphere, it may auto-ignites depending on the hydrogen jet and surrounding geometrical conditions. The present work will clarify the high pressure hydrogen jet auto-ignition phenomena after it spouting from a hole or a tube experimentally using a small shock tube and a numerical analysis. It is shown that high pressure hydrogen jet leaking from a small hole does not auto-ignite and that coming out from tube it auto-ignites. Some numerical calculations are also presented to explain the experimental evidences. The parameters for such auto-ignition depend on hydrogen pressure, tube diameter, and tube length. A discussion of the limit of these parameters is also presented.

I. Introduction

HYDROGEN is one of the mostly paid attention fuels as an alternative one of petroleum resources. However since hydrogen has a property of a wide flammability limit and a low ignition energy, there need a standard for hydrogen safety for its utilization. In this point of view basic and applied research has been started worldwide recently. The very original study has been performed by Wolanski and Wojcicki¹ (1973) using a small shock tube system to produce a hydrogen jet into an atmosphere. The piston inside the shock tube is droved by detonation and the piston-compressed hydrogen breaks a copper plate to produce a high pressure hydrogen jet to atmosphere. Tanaka et al.² showed experimentally the ignition of hydrogen jet without any ignition sources. They fill up a tube by high pressure hydrogen directly to break a copper plate and to release it into atmosphere. Recently Dryer et al.³ performed several experiments with high pressure hydrogen jets spouting to atmosphere by using compressed hydrogen bottles and burst disks to get auto ignition and diffusion flame. Liu et al.⁴ studied numerically the high pressure hydrogen jet spouted into atmosphere using a direct numerical simulation method with a full hydrogen chemistry. The computational results coincide with the experimental ones qualitatively.

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A hydrogen jet ignition mechanism at atmosphere is as following: (1) when high pressure hydrogen jet is spouted into atmosphere, a shock wave is formed in front of the hydrogen jet; (2) this head shock heats up the air behind it by compression up to more than 1000 K depending on the hydrogen jet pressure; (3) the hot heated air exchanges mass and heat with the cold expanded hydrogen jet at its contact surface through mass and heat diffusion to lead to local ignition. Few experimental studies of high pressure hydrogen auto-ignition have been performed due to the difficulty of using such high pressure hydrogen. The present study uses the shock tube technique to get a high pressure hydrogen jet and to clarify its ignition mechanism experimentally and numerically. And when the extended tube is used, the relation among tube diameter, tube length, and hydrogen jet pressure will be discussed.

II. Experimental Method

Experiments are performed to use a small shock tube with and without extended tube. Two different sizes of extended tubes are used to see the extended tube effect for hydrogen jet auto-ignition. A Shimadzu high speed camera is used to observe the shock wave and contact surface location and auto-ignition. OH emission for auto-ignition cases are observed using an ICCD camera (La Vision) with a five nano second aperture time.

A. Small Shock Tube System

It is quite difficult to do an experiment using high pressure hydrogen in a university laboratory level. From one of the reasons the present experimental system is set up using a small shock tube to compress hydrogen with shock wave. This system was used by Wolanski and Wojcicki¹ in 1979. After high pressure hydrogen ruptures a copper plate, it is released to a large dump tank without any leak from tubing and any noise from the system.

Figure 1. Schematics of experimental system

Figure 1 shows the present experimental system. The shock tube has a high pressure section which is filled up with 8 MPa of nitrogen and a low pressure section which is filled with 0.9 MPa of hydrogen. A diaphragm is installed downstream the low pressure section, which thickness depends on the jet pressure condition of 3.8 MPa and 6.8 MPa. The further downstream of the diaphragm is the test section of dump tank. The piston can be used in this shock tube where the piston compresses hydrogen instead of nitrogen. The hole and tube diameters are two kinds; 4.8 mm and 10.0 mm. The tube length is 18 through 180 mm depending on the connecting condition.

Two observation windows of 200 mm in diameter are installed at the test section to see whether hydrogen ignites or not and to take a Schlieren pictures, movies, and OH emission profile data. Hydrogen in the dump tank is exhausted to outside through a exhaust pipe. The diaphragm used are PET films of 25, 38, 50 μ m thick and sometimes a couple of PET films are put together for the higher pressure case. A PCB pressure transducer is installed just behind the diaphragm to measure the pressure just after the diaphragm is ruptured. A high speed

camera is used to take the sequence of the fast phenomena which trigger is taken from the pressure taken from the pressure transducer just behind the diaphragm. The maximum speed of the frame of the high speed camera is one million frames per second.

The whole measurements are controlled and measured by LabVIEW system (National Instruments). Since a very short time operation is necessary, the LabVIEW system can support the time triggering system for high speed valve system and high speed camera system.

B. Non-extended tube (a hole)

Instead of using a hole the very short tube is utilized as a hole. The detailed condition of this case will be different from the typical hole, but its essential role will be the same. The size of the hole is 4.8 and 10.0 mm in diameter.

C. Extended tube

In the present study three different cases are investigated; the case without tube, the case with a tube of 10 mm in diameter, and the case with a tube of 4.8 mm in diameter. The tube length will be 48, 71, 113 mm for 4.8 mm in diameter (Fig. 2) and 50, 100, 180 mm for 10 mm in diameter (Fig.3). Both the straight tube and the combined tube have a smooth inner surface.

Figure 2 The tube of 10 mm inner diameter: straight tube.

Figure 3 The tube of 4.8 mm inner diameter: combined tube.

D. Experimental conditions

Two different major cases are performed for the present study; the case without tube and that with tube. These cases will provide us the information of the role of tube.

(1) The case without extended tube

The case without tube corresponds to the case that a hole is suddenly opened at a high pressure hydrogen tank and a high pressure hydrogen jet is spouted into atmosphere. The high speed movie of the jet is taken using the Schlieren method and a Shimazu camera. The experimental condition for this case is that the diaphragm used is a 50 μ m PET film, which provides the rupture pressure of 3.8 MPa and the hydrogen pressure of 1.6 MPa at the hole exit or tube exit. The air pressure in the dump tank is 0.1013 MPa, the same as standard atmospheric pressure. The pressure ratio, p_4/p_1 , of the shock tube relation in this case is 38, where p_1 is the dump tank initial pressure and p_4 is the compressed hydrogen pressure. In order to check the higher hydrogen pressure case, the dump tank pressure is reduced to 0.0113 MPa, which corresponds to the pressure ratio of 336 using the same rupture pressure of 3.8 MPa.

(2) The case with extended tube

Two kinds of tube are used to this experiments; straight and combined tube. However both tubes are straight and only their sizes are different. The dimensions of these tubes are: one is 10 mm in diameter and 50, 100, 180 mm in length and another is 4.8 mm in diameter and 48, 71, 113 mm in length as stated in the B. The hydrogen pressure and atmospheric air pressure are 0.7 MPa and 7.0-8.0 MPa depending on the rupture. In order to take a picture of ignition, the pressure gauge is used for not only to measure local pressure, but also to provide a trigger signal to the high speed camera.

III. Numerical method

It is assumed that a high pressure hydrogen jet is injected into the still air through a small hole or tube from a high pressure hydrogen tank. The total pressure of hydrogen is 3.8 or 6.8 MPa, which is a little different from the experimental condition and the diameter of the hole is 10 mm or the tube diameter of 10 mm.

A. Governing equations

The governing equations are the compressible two-dimensional axisymmetric Navier-Stokes equations, the conservation equations of total energy and chemical species, and the equation of state. The equation of total mass conservation is solved additionally. A model by Petersen and Hanson⁵ with 9 species (H_2 , O_2 , O , H , OH , HO_2 , H_2O_2 , H_2O , and N_2) and 18 reactions is used to solve the present problem. This reduced kinetics mechanism has good performance of ignition delay time and heat release within a wide pressure range from 1 to 600 atm. The air is assumed to be composed of 22% O_2 and 78% N_2 in volume. The diffusion flux is evaluated using Fick's law with binary diffusion coefficients. The transport coefficients of each chemical species; viscosity, heat conductivity, and binary diffusion coefficient, are evaluated using the Lennard-Jones intermolecular potential model⁶, and those of the gas mixture are calculated by Wilke's empirical rule⁷. The enthalpy of each chemical species is derived from NIST data base⁸. The buoyancy, bulk viscosity, Soret and Dufour effects are neglected.

B. Computational scheme

The governing equations are discretized in a finite difference formulation. The convective terms are evaluated using the second-order Explicit Harten-Yee Non-MUSCL modified-flux type TVD scheme, considering the properties of the hyperbolic equations. The viscous terms are evaluated with the standard second-order central difference formulae. The time integration method is the second-order Strang-type fractional step method. The chemical reactions are treated by a point implicit method to avoid the stiffness.

E. Initial conditions

The total pressure of the hydrogen in the tank is 3.8 or 6.8 MPa and the total temperature is 300 K. The diameter of the hole is 10 mm and that of tube is also 10 mm. The jet is choked in the hole and its Mach number is unity, i.e. it has a static pressure of 21.13 MPa, a static temperature of 250 K and a velocity of 1220 m/s. But sometimes it does not choke at the exit of the hole or tube. The inner surfaces of the hole are assumed to be slip walls and the hydrogen jet has a uniform velocity profile. The jet exit is on the same plane as the outside surface of the wall. These conditions are kept in the simulation. The computational domain is a rectangular zone with a length of 80-90 mm in x-direction and a width of 80-120 mm in y-direction. The x-direction is the jet axial direction. The left boundary is a solid wall with the hole located at its center. Non-slip conditions are imposed on the wall and the free stream conditions are applied to the other three boundaries. The flow is adiabatic. At the initial state, the computational region is filled with still air at 1 atmosphere and 300 K. No artificial disturbance is imposed. The grid system is rectangular and a uniform grid size of $dx = dy = 20 \mu m$ is accepted. The reason is that the species profiles are extremely narrowed by such a high pressure hydrogen jet and the flame front becomes very thin⁹. It is estimated that this grid spacing is close to the Kolmogorov scale according to the references^{10, 11}. The total grid number is 27 million.

IV. Results and discussion

Experimental and numerical studies have been performed to see the physics and its detail of hydrogen jet ignition. Experimentally high pressure hydrogen jet is spouted from a hole of 4.8 and 10.0 mm in diameter and two kinds of extended tube of the same diameter. The tube length is changed from 18 to 180 mm. Numerical condition is a little different from the experimental ones; the hole and extended tube diameter is 8 mm and the initial hydrogen pressure is 3.8 MPa which is close to the experimental conditions.

A. the case of hydrogen jet spouted from a hole

The physics of high pressure hydrogen spouted from a hole has been studied by Liu et al.^{4, 12} that when the high pressure hydrogen jet coming out of the tank, a head shock wave forms in front of hydrogen jet, which strength depends on its strength of the jet. The leading shock wave heat up air behind it by its compression effect and provides an ignition at the hydrogen jet front, where there is a mixing between hot air and hydrogen. The reaction front at the contact surface between hydrogen and air is thin and becomes weak and extinguishing as the leading

shock wave goes further downstream. The numerical study by Liu et al.^{4,12} showed that there is no steady ignition when the leading shock wave propagates downstream at the initial hydrogen jet pressures of 10 through 70 MPa.

The experimental work for the case of hydrogen jet spouted from a hole is observed at the low hydrogen jet case by high speed Schlieren movie as shown in Fig. 4. The pictures showed no ignition which is quite different from the ignition case shown later. The leading shock wave is spherical and the distance between the leading shock and the contact surface becomes apart as the shock wave propagates, but the time passed, this distance becomes shorter again; the contact surface approaches to the leading shock front together with compression waves generated by the hydrogen jet. Then the shock front becomes thicker as time passed. Besides the hydrogen jet behind the leading shock wave becomes more turbulent.

Figure 4 High pressure hydrogen jet from a hole of 10 mm in diameter at the hydrogen pressure of 2.3 MPa.: no ignition case at the time of (a) 40 μ s, (b) 72 μ s, (c) 168 μ s, (d) 248 μ s, (e) 288 μ s, (f) 328 μ s from the time at the exit of the hole.

This case is also confirmed by a numerical simulation. Figure 5 shows the similar case as the experiment shown in Fig. 4. The numerical initial condition for this case is that the hydrogen temperature, pressure, and Mach number are 671.4 K, 3.8 MPa, and 0.5 (the case is not choked at the tube exit.), respectively and the atmospheric air temperature and pressure are 300 K and 0.1 MPa. The calculation does not include the chemical reaction since the hydrogen temperature at the hole is low enough to auto-ignite. As shown in Fig. 5-(a), the numerical shock wave front propagates faster than that of experiments because the pressure of the experiment is lower than that of the numerical one. We could not measure it at the hole exit at this time. The experimental hydrogen pressure of 2.3 MPa comes from the shock tube relation. Figure 5-(b) shows the numerical temperature history at the hole exit. The temperature suddenly decreases at around 180 μ s due to the contact surface.

Figure 5 The location of the leading shock wave and temperature at the hole for the case of hydrogen jet spouted from the hole of 10 mm in diameter, its pressure at the hole of 3.86 MPa and its temperature at the hole of 671.4 K comparing with the experimental hydrogen Pressure of 2.3 MPa.

The important parameters for high pressure hydrogen jet spouting from a hole of the high pressure tank are hydrogen pressure and hole size; diameter and length. The previous studies^{4, 12} show that when the hydrogen pressure becomes high, the leading shock wave in front of the hydrogen jet becomes strong, but the jet expansion effect also becomes strong to weaken the jet faster than the case of lower pressure hydrogen jet and the hydrogen jet will not auto-ignite at the hydrogen pressure as high as 70 MPa. When the hole diameter becomes larger, the jet becomes weaker to keep the chemical reaction at the contact surface and the auto-ignition will not occur.

B. the case of hydrogen jet spouting from a tube

When hydrogen spouts from a tube, the physics of high pressure hydrogen spouting is quite different from the one spouting out of a hole. When the high pressure hydrogen propagates in the tube after it bursts a diaphragm, a leading shock wave arises in front of the hydrogen in the tube. This leading shock wave does not decay due to the confined walls. Then the air behind the leading shock wave becomes very hot and ignites the hydrogen in the tube. The boundary layer may be developed after the leading shock wave to mix the hot air and hydrogen more effectively. The hot ignited mixture comes out of the tube to keep the flame without extinguishing.

Figure 6 Sequential OH emission photos of high pressure hydrogen jet of 2.3 MPa through the tube at 4.8 mm in and 115 mm in length: (a) 125 μ s after bursting the diaphragm, (b) 150 μ s after bursting the diaphragm, (c) 175 μ s after bursting the diaphragm, (d) 200 μ s after bursting the diaphragm.

Figure 7 High pressure hydrogen jet spouting from the tube of 4.8 mm in diameter and 116 mm in length at the hydrogen pressure at the exit of 6.8 MPa. The jet is auto-ignited at the beginning of spouting out of the tube: (a) 20 μ s, (b) 84 μ s, (c) 148 μ s, (d) 196 μ s, (e) 276 μ s, (f) 328 μ s.

The experiments are conducted using several types of tube. The hydrogen jet ignition condition is studied for the parameter of tube diameter, tube length, and hydrogen pressure. The higher hydrogen pressure gives its auto-ignition easier. Figure 6 shows the sequential OH emissions of the hydrogen ignition at the condition of 4.8 mm diameter tube with the length of 115 mm. and the hydrogen pressure of 6.8 MPa. The OH emission photos show that the flame stays at the exit of the tube. In this case the emission strength becomes weak as time passes due to the burning out of hydrogen.

The high speed Schlieren movie for the case of auto-ignition is shown in Fig. 7. The hydrogen jet configuration in this case is quite different from the one shown in Fig. 4. The hydrogen jet in Fig. 7 is already auto-ignited at the exit of the tube (probably it is ignited even in the tube.) and its turbulent flame propagates nearly cylindrically into the atmospheric air due to the jet propagation direction. In this case the leading shock wave does not help for the ignition, but it does for keeping the jet flame. At the last three pictures (Figs. 7-(d), (e), (f)) do not show their difference since hydrogen in the tube is almost burned, which is the similar to the case in Fig. 6. In these photos the local burnings are observed by the bright emissions. However the shock wave front velocities of the experiment for the ignition case using the 4.8-mm-in-diameter tube and for the non-ignition case using the 10.0-mm-in-diameter tube are similar along the flow direction.

The numerical results for high pressure hydrogen spouting from a hole or tube of 10 mm in diameter at the hydrogen pressure of 3.8 MPa are shown in Fig.8 where (a) is the leading shock front location from the hole or tube exit to the time after bursting the diaphragm and (b) is the velocity of the leading shock front vs the time after bursting the diaphragm. The results show clearly that the leading shock front decays quickly for both cases within 100 μ s. Both are the cases without auto-ignition, which corresponds to the experimental results for the case of hole and the case of tube with 10 mm in diameter. Although the present tube case does not have auto-ignition, the confined effect of tube shows the stronger leading shock wave and longer time to keep its strength.

Figure 8 A comparison between hydrogen jet spouting out from a hole and from a tube of 10 mm in diameter at the hydrogen pressure of 3.8 MPa: (a) leading shock position and (b) leading shock velocity.

In the case of tube the smaller diameter tube and the higher hydrogen pressure provide the hydrogen auto-ignition as shown in Table 1. From the table the hydrogen auto-ignition will occur at higher hydrogen pressure, larger tube diameter, and longer tube length. There should be the hydrogen pressure limit under 3.8 MPa, the tube diameter limit smaller than 4.8 mm, and the tube length limit shorter than 71 mm.

Table 1 Auto-ignition conditions for hydrogen pressure, tube diameter, and tube length

Hydrogen pressure [MPa]	Tube diameter [mm]	Tube length [mm]	Ignition
2.7	4.8	71	no
3.8	4.8	48	no
3.8	4.8	71	yes
3.8	4.8	113	yes
6.8	4.8	48	no
6.8	4.8	71	yes
6.8	4.8	113	yes
6.8	10.0	118	no
6.8	10.0	180	yes

V. Summery and conclusion

High pressure hydrogen jet auto-ignition by spouting from a hole or various configurations of tube was studied experimentally and numerically for the hydrogen safety point of view. Experimentally a 5 nano second ICCD camera and high speed movie are used to observe the leading shock wave, hydrogen jet, mixing, and auto-ignition. Numerically two-dimensional axisymmetric NS equations are integrated to see the leading shock wave, hydrogen jet, and mixing behaviour.

Two different conditions are investigated for the auto-ignition of hydrogen jet spouting from the high pressure tank: one is from a hole and the other is from a tube. The hydrogen jet coming out from a hole does not auto-ignite at the pressures high up to 700 K for any size of hole diameter. The different hole condition from the one we made may provide such ignition if such high pressure jet experiment could be performed.

When the high pressure jet is coming out from the tube, the story is quite different from the one coming out of a hole. The leading shock wave coming out of the hydrogen front becomes strong in the tube and the air between this

shock wave and the hydrogen front is heated up by the shock wave compression. Through the mixing at the contact surface between air and hydrogen in the tube, there occurs an auto-ignition in the tube. Then the flame coming out of the tube ignites the hydrogen jet at the atmosphere. This ignition at the atmosphere is confirmed by both the ICCD camera and the high speed movie.

The auto-ignition condition for hydrogen jet spouted from the tube is the higher hydrogen pressure, smaller tube diameter, and longer tube length. There are limits for these conditions and the present study has not confirmed in detail yet. But the hydrogen pressure limit could be lower than or as low as 3.8 MPa, the tube diameter could be smaller than 4.8 mm, and the tube length could be 71 mm.

The further detail experiments and numerical simulation will be performed in the future.

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