MECHANISM OF HIGH PRESSURE HYDROGEN AUTO-IGNITION WHEN SPOUTING INTO AIR

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ABSTRACT

High pressure hydrogen leak is one of the top safety issues presently. This study elucidates the physics and mechanism of high pressure hydrogen jet ignition when the hydrogen suddenly spouts into the air. The experimental work was done elsewhere, while we did the numerical work on this high pressure hydrogen leak problem. The direct numerical simulation based on the compressible fluid dynamics considering viscous effect was carried out with the two-dimensional axisymmetric coordinate system. A detailed model of hydrogen reaction is applied and a narrow tube attached to a high pressure reservoir is assumed in the numerical simulation. The exit of the tube is opened in the atmosphere. When high pressure hydrogen is passing through the tube filled by atmospheric air, a strong shock wave is formed and heats up hydrogen behind the shock wave by compression effect. The leading shock wave is expanded widely after the exit, hydrogen then mixed with air by several vortices generated around the exit of the tube. As a result, a couple of auto-ignitions of hydrogen occur. It is found that there is a certain relationship between the auto-ignition and tube length. When the tube becomes longer, the tendency of auto-ignition is increased. Additionally, other type of auto-ignitions is predicted. An explosion is also occurred in the tube under a certain condition. Vortex is generated behind the shock wave in the long tube. There is a possibility of an auto-ignition induced by vortices.

1.0 INTRODUCTION

Recent environmental and energy crises push up a change of energy source from the fossil fuel to others. Hydrogen is one of the possible candidates for such energy source. However, there are some problems when we use the hydrogen energy safely and efficiently. One of the serious problems is that hydrogen might lead to an accidental explosion. High pressure tanks or reservoirs are often used to store hydrogen in many practical purposes. It is necessary to compress hydrogen gas to decrease its volume since the hydrogen energy density is low.

It is well known that the accidental release of hydrogen from a high-pressure tank into the air can produce a strong shock wave which heats up air to a high temperature to ignite hydrogen. The auto-ignition of hydrogen leads to an explosion under a certain condition. From the safety point of view, this problem is important in practical cases such as a pressure vessel containing high-pressure hydrogen or an automobile fuel cell, etc.

Many studies have been performed to clarify the mechanism of an explosion induced by high pressure hydrogen since 1973 when Wolański and Wójcicki [1] did this study using a shock tube system to find out the auto-ignition structure at the exit of the tube using a Schlieren photographic system. After their study, Tanaka et al. [2] performed a similar experiment to confirm the ignition in 1979. Before the extensive work on the auto-ignition problem, the related work was performed by Uejima et al. [3, 4] to see the turbulent effects on an ignition of hydrogen free jet.

For the last few years, studies about high pressure hydrogen jet spouting from a tube have been done extensively to clarify the relation among tube diameter, tube length, hydrogen jet pressure, and so on. Liu et al. [5] and Bazhenova et al. [6] calculated the jet coming out to the air at the room temperature, where a sudden expansion occurred near an exit wall to reduce it to a very low temperature. Dryer et al. [7] demonstrated spontaneous ignition (auto-ignition/inflammation and sustained diffusive combustion) by sudden compressed hydrogen releases from a tube, but the details of the experimental conditions were unknown. Golub et al. [8], Mogi et al. [9] and our group [10] presented a relationship between inlet pressure of hydrogen and length of a tube. It is mentioned that the auto-ignition using a long tube occurs easier than that using a short one. Xu et al. [11] mentioned that when the tube was sufficiently long under certain release pressure, an auto-ignition would initiate inside the tube at the contact surface due to mass and energy exchange between low temperature hydrogen and shock-heated air through molecular diffusion.

In the present work, a direct numerical simulation (DNS) with a detailed chemical model is performed to clarify the ignition mechanism of high pressure hydrogen jet spouting from a narrow tube connected to the large hydrogen reservoir. Especially, we notice the phenomena of hydrogen explosion in the tube.

2.0 METHOD

2.1 Governing Equations

The direct numerical simulation based on the compressible fluid dynamics considering viscous effect is performed to clarify the explosion mechanism of high pressure hydrogen. The governing equations for this study are the conservation equations of mass, momentum, energy, and chemical species together with the equation of state.

For chemical reaction model, the reduced kinetic mechanism [12], including nine chemical species (H_2, O_2, O, H, OH, HO_2, H_2O_2, H_2O, and N_2) and 18 elementary reactions, is used to solve the present problem. This mechanism has a good performance for ignition delay time and heat release within a wide pressure range from 0.1 to 60 MPa. The diffusion flux is evaluated using the Fick's law with binary diffusion coefficients. The transport coefficients of each chemical species, namely, viscosity, heat conductivity, and binary diffusion, are evaluated using the Lennard–Jones intermolecular potential model [13], and those of the gas mixture are calculated by the Wilke's empirical rule [14]. The buoyancy, bulk viscosity, and Soret and Dufour effects are neglected.

2.2 Computational Scheme

The governing equations are considered on the (r, z) plane under the assumption of axial symmetry for the cylindrical coordinate system. These equations are given in discrete forms with a finite difference method. 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 the point implicit method to avoid stiffness.

2.3 Flow Field Model

Figure 1 shows a schematic of the flow field used for the present numerical simulation. It is assumed that high-pressure hydrogen provided from a narrow tube is injected into the still air. The tube is connected to the large hydrogen reservoir at the left-hand side in the figure. The diameter of the tube is 4.8 mm, while the length of the tube is varied. The initial condition of the whole region is set to be the atmospheric condition. To save the CPU cost, the numerical domain is extended gradually along z-axis with the propagation of shock wave induced by high pressure hydrogen.

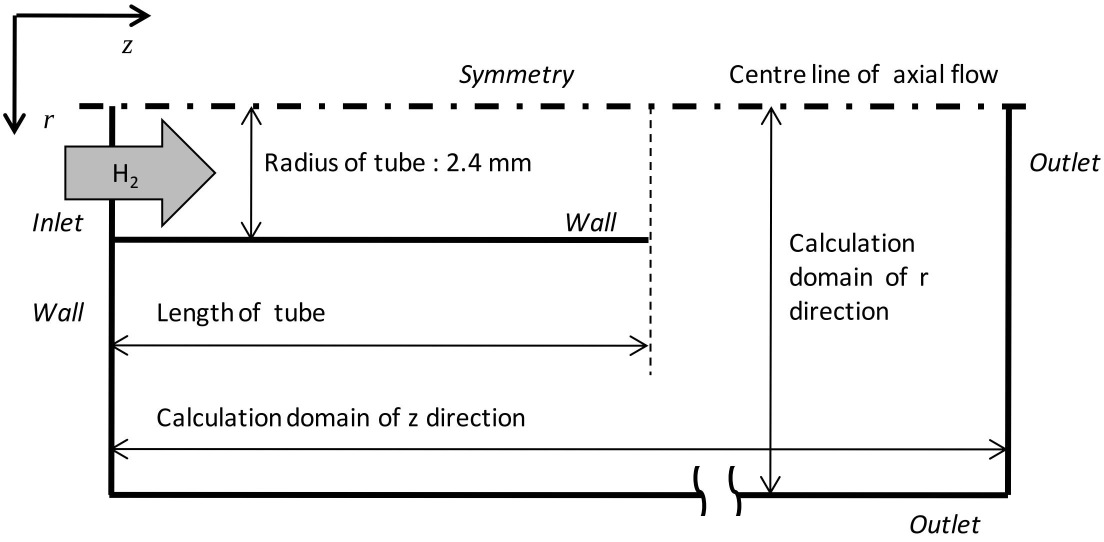


Figure 1. Analytical model and boundary condition of numerical simulation

2.4 Boundary Conditions

As shown in Fig. 1, there are six boundaries for the present calculation: an inlet boundary, two wall boundaries, two outlet boundaries, and a symmetry boundary. At the inlet boundary, high pressure hydrogen, which velocity profile is given by a uniform value, is provided. The inlet pressure and temperature are 21.1 MPa and 507 K, which are estimated by the choked condition. The reservoir pressure corresponds to 40.0 MPa. The tube wall and left-hand wall have nonslip wall condition. The free stream condition is applied to two outlet boundaries.

3.0 RESULTS AND DISCUSSION

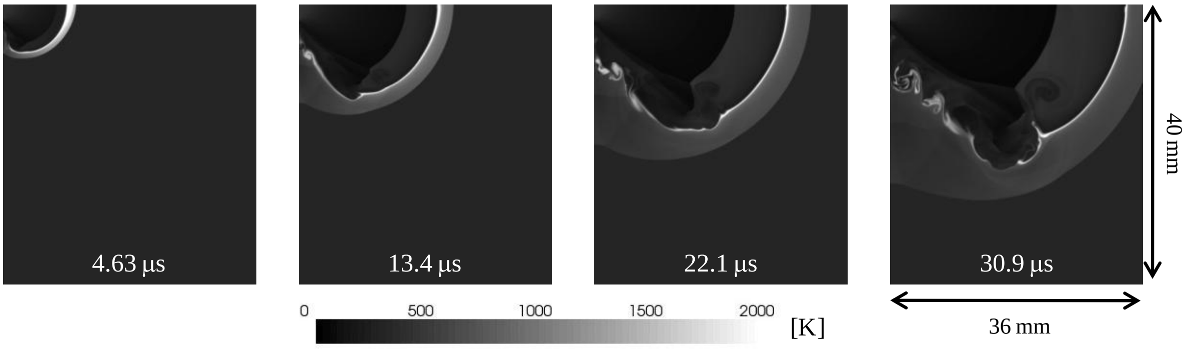
3.1 Effect of Tube Length

To investigate the effect of tube length, four cases listed in table 1 are calculated. The length of tube is varied and numerical domain is extended along z-axis depending on the tube length. The radius of the domain is fixed at 40 mm for all cases. The longer tube case has a wider calculation domain to z-axis. The grid width for all cases is set to be 20 μm uniformly, which is close to the Kolmogorov scale according to the references [15, 16]. The case #1 is the no tube case, i.e., hydrogen jet spouts from a small hole on the reservoir wall. The case #2 is the short tube case. On the other hand, the cases #3 and #4 use a slightly long tube.

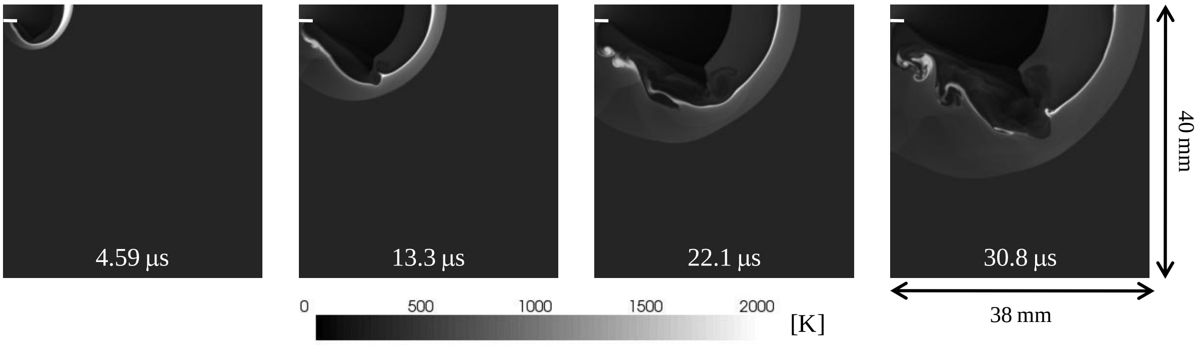
Table 1. Condition of each case.

#	Length of tube [mm]	Calc. domain (r × z) [mm]	Grid
1	0	40 × 36	3,600,000
2	2	40 × 38	3,800,000
3	10	40 × 46	4,600,000
4	20	40 × 56	5,600,000

Figure 2 shows the sequential temperature distributions for the cases of #1 and #2. The time in μs shown in the figure indicates the time after the hydrogen gas begins to break into the calculation domain from the inlet at the upper left corner. The white horizontal line fixed in the upper left corner shown in Fig. 2-(b) indicates the tube wall. The shock wave is generated immediately after the hydrogen breaks into the numerical domain in both cases, and hydrogen gas begins to expand semi-spherically in the still air. The shock wave heats up the air to a high temperature of over 2000 K which has enough energy to induce hydrogen auto-ignition under the atmospheric condition. The mixing of hydrogen with air is promoted by vortices generated around the side area of the exit. This mixture layer at high temperature leads to a strong chemical reaction. As a result, an auto-ignition may occur in this area. It seems that vortices in the case #2 grow up a little more complicated than those in the case #1. In the case #2, there is a little more space around the exit. Therefore, vortices are generated easily near the exit.



(a) Case #1



(b) Case #2

Figure 2 Temperature distributions for cases #1 and #2

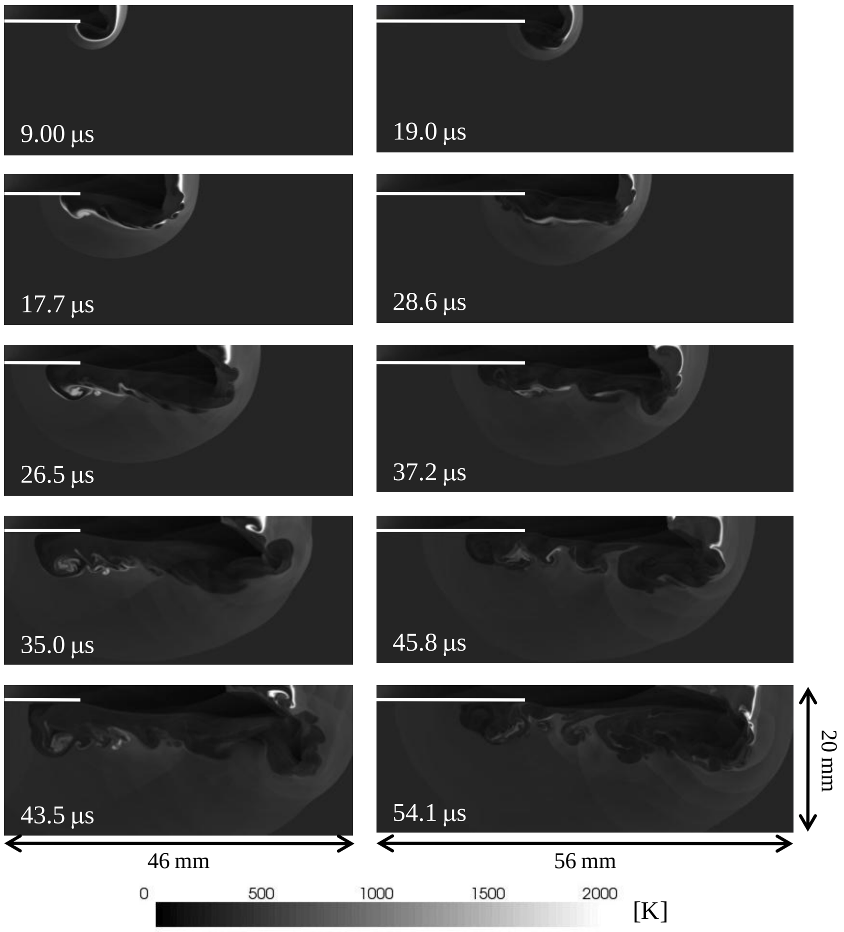
For a longer tube case, the cases #3 and #4 are calculated. It is found that there is a clear difference compared with the results shown in Fig. 2. The shock wave does not expand semi-spherically around the tube exit. In the side region of the tube, many vortices are generated, and the mixing of hydrogen with air is promoted efficiently by the vortices. However, temperature becomes low around the side region of the tube due to the expansion. The high-temperature area around there disappears with time. Therefore, it is recognized that the chemical reaction is not held in the region.

On the other hand, the high-temperature area is maintained behind the shock front. The energy of the shock wave keeps high temperature. Some vortices are generated there, which are not appeared in the cases #1 and #2 shown in Fig. 2. The vortex generated near the shock front plays a role in mixing the hydrogen provided from the tube with the surrounding air. It tends for the chemical reaction to be maintained easily. Hence, the auto-ignition of hydrogen front of the shock wave is maintained for a while.

Figure 4 shows the maximum temperature history of all 4 cases until high temperature area at contact surface reaches to the outlet boundary. For all cases, the maximum temperature starts from ca. 3250 K, and then gradually decreases. For the longer tube cases, #3 and #4, the decreasing rate of the maximum temperature until ca. 8 μs is more remarkable than that of cases #1 and #2. From Fig. 3-(a), it is found that the shock wave starts to expand at 9.00 μs. It means that decreasing rate of the maximum temperature in the tube is larger than when the shock wave expands the ambient air.

The maximum temperature is gradually decreasing until ca. 28 μs for the cases #1 and #2. Although the maximum temperature slightly rises at 30 μs, the temperature is gradually decreasing again. The mixing layer generated near the exit induces an auto-ignition of hydrogen, however, enough energy and mixture to keep reaction are not generated after the ignition.

As for the case #3, when the shock wave exits the tube, the temperature slightly rises. As for the case #4, the temperature in the tube is almost constant at ca. 2600 K after 10 μs. When the shock wave exits the tube, the temperature slightly rises too in this case #4. For Both cases, after the shock wave exits the tube, the maximum temperature oscillates around 2400 K. Therefore, it is found that an auto-ignition of hydrogen is maintained for a while.



(a) Case #3

(b) Case #4

Figure 3 Temperature distributions for cases #3 and #4

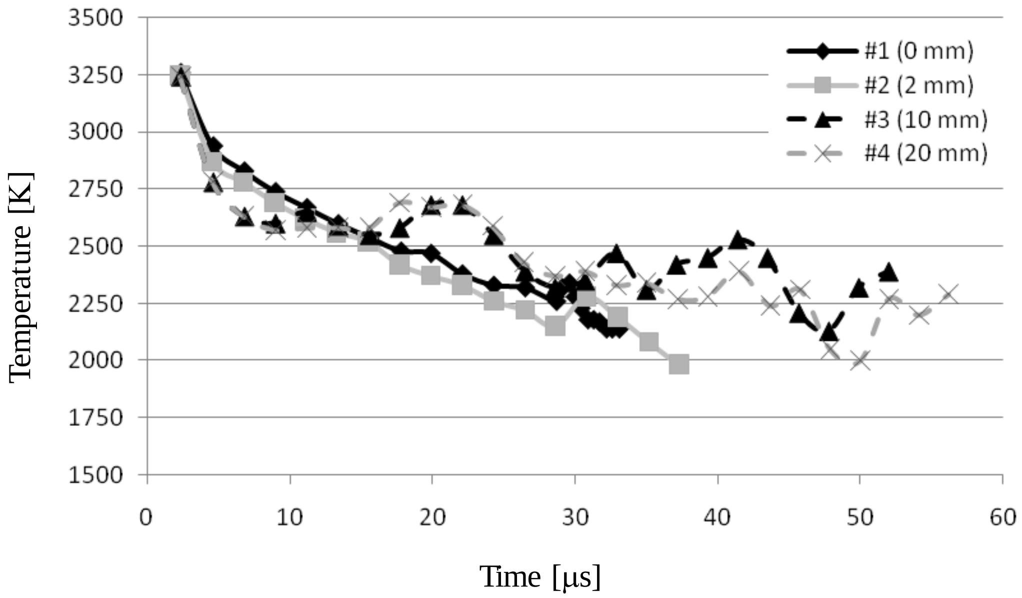


Figure 4 Maximum temperature history of each case

3.2 Phenomenon inside a long tube

To discuss about the phenomenon inside a long tube, a finer grid is applied to the calculation. The grid width is set to be 10 μm along z-axis and 5 μm along r-axis to resolve vortices in the Kolmogorov scale with an accuracy. In this calculation, inside the tube is only solved to save the CPU cost. The inlet pressure is set to 41 MPa to comprehend the effect of the pressure greatly.

Figure 5 shows the sequential temperature distributions in the long tube. From the left side of the figure, the high pressure hydrogen spouts into the tube. The temperature of hydrogen gas rapidly increases between the shock wave and contact surface. At first, the shape of the contact surface is spherical, and then it gradually becomes a flat shape. When the shock wave passes at ca. 30 mm, the shape of the contact surface is changed significantly. This spherical shape having a large curvature is formed near the tube wall at the contact surface. While, it seems that the contact surface near the centre axis is started to break. The high temperature area over 2000 K is formed there, and grows up widely. It is thought that the strong chemical reaction occurs there.

To investigate the phenomena inside the tube in detail, the temperature contours around the shock wave at 22.9 μs is shown in Fig. 6. The bottom contour is an enlargement figure in the part enclosed by the dotted line in the upper figure. The lateral line which separates two waves is recognized apparently in the upper figure. The line gradually moves out from the wall, crosses the centre axis, and moved toward the wall. After reaching the wall, the line reflects at the wall, and gradually traveled away the wall again. In front of the line, many contour lines are formed densely. As for the enlargement figure, the complicated flow structure is formed behind the contact surface. It is expected that the complicated flow structure mixes hydrogen with air and that it causes a strong chemical reaction easily.

Figure 7 shows pressure contours at 7.4 and 22.8 μs. As for the upper figures, the light area shows a higher pressure, while the dark area shows a lower pressure. The maximum pressure is 15.0 MPa at 7.4 μs. Although the maximum pressure at 22.8 μs is decreasing, more complex structure appeared as shown in the bottom figure at 22.8 μs. It is also found that a kind of whisker in the length of ca. 0.5 mm appeared apparently at 22.8 μs. One of the whiskers connects the centre of a pressure vortex.

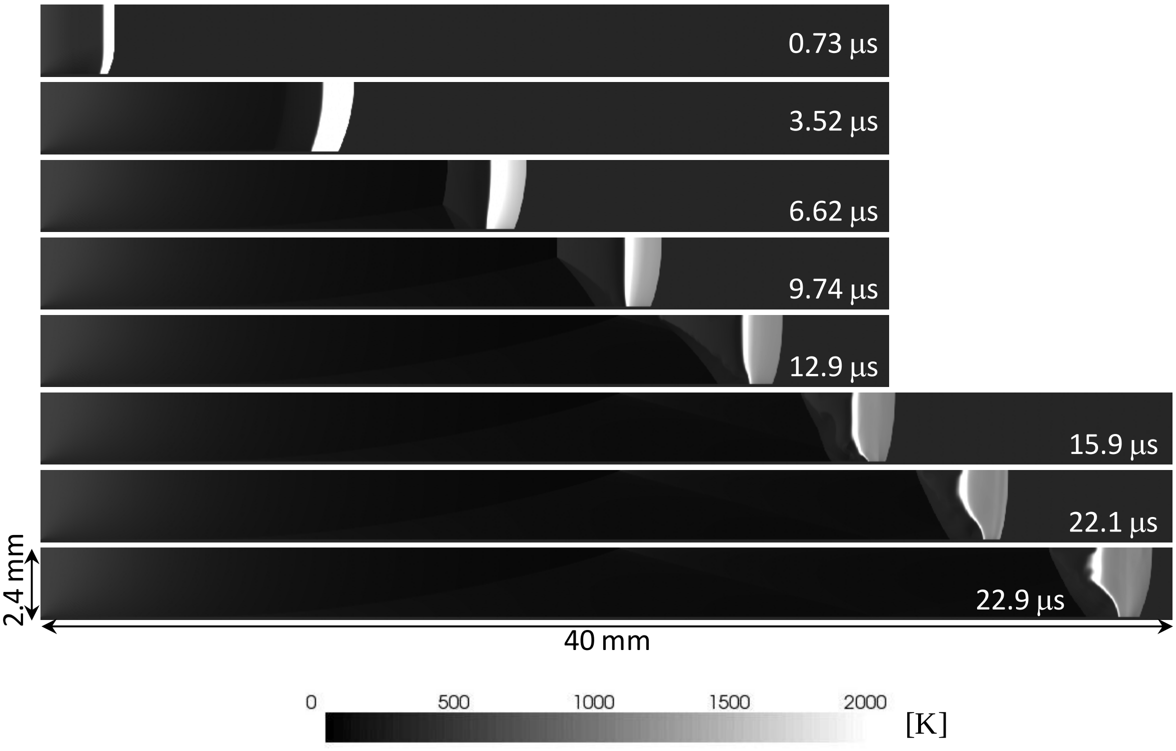


Figure 5 Temperature distributions in the long tube

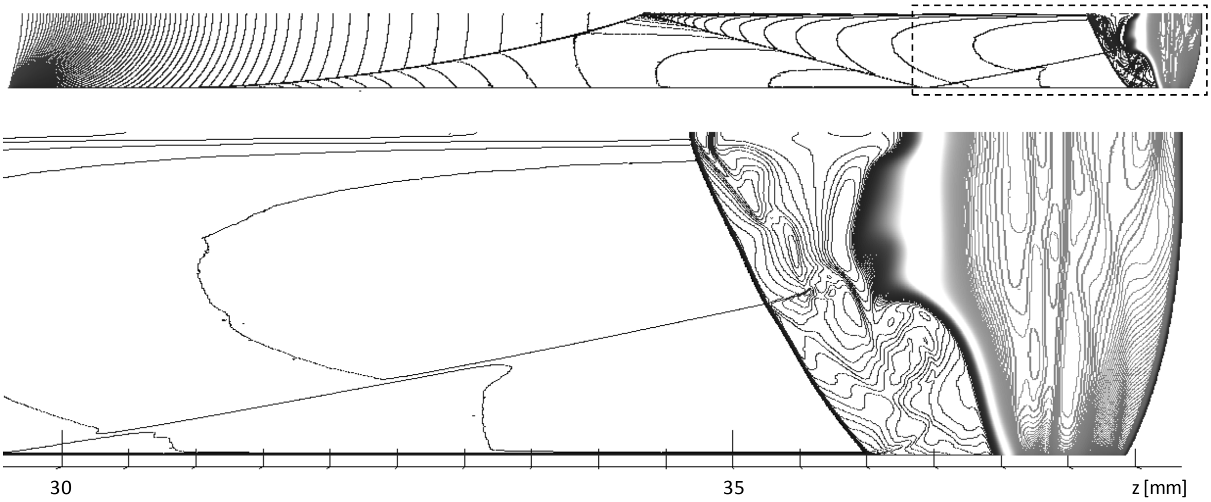
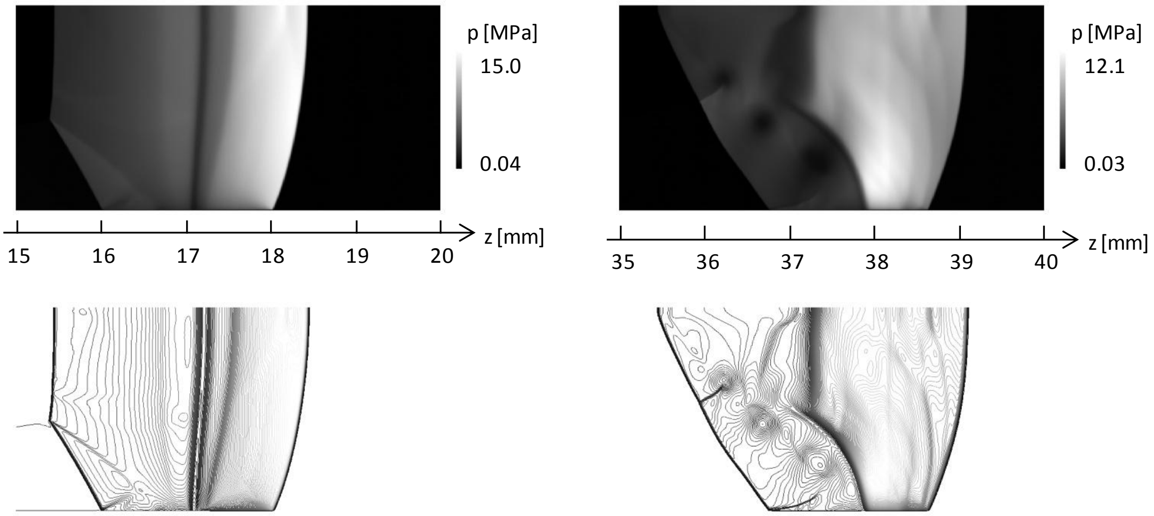


Figure 6 Temperature contours in the tube and around the contact surface at 22.9 μs

To make sure the existence of vortices, the vorticity distribution is shown in Fig. 8. Three vortices in line are shown in this figure. This may cause an effective mixing between hydrogen and shock-heated air, and then an auto-ignition of hydrogen inside the tube may occur.



(a) 7.4 μs

(b) 22.8 μs

Figure 7 Pressure contours in the tube and around the contact surface

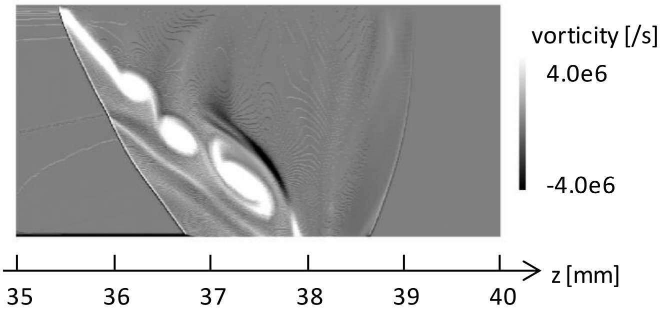


Figure 8 Vorticity distribution in the tube and around the contact surface at 22.8 ms

4.0 CONCLUSIONS

In this paper, the high pressure hydrogen jet spouting into the air is reviewed and is shows some recent results with a direct numerical simulation. Two types of calculation are performed to analyze two types of auto-ignition. One is for solving the auto-ignition phenomenon outside of the tube, and another one is for solving that inside of the tube.

As for the outside of the tube, there is a relationship between auto-ignition and length of the tube. When the tube becomes long, there is an enough space to mix hydrogen and surrounding air around the exit. Under such a condition, many vortices are generated, and high temperature area tends to be maintained. Therefore the auto-ignition is promoted further. It is found that the space near the exit is important to induce the auto-ignition of hydrogen. To avoid the auto-ignition, it is necessary to reduce the space near the exit.

As for the inside of the long tube, the shape of the contact surface is gradually changed while the leading shock propagates. The spherical shape of contact surface in the early time becomes a flat shape, and then a spherical shape having a large curvature is formed near the tube wall at the contact surface. The complicated flow structure with high temperature is formed behind the contact surface. Three vortices in line are also found at 22.8 μs. The complicated structure may cause an effective mixing between hydrogen and shock-heated air, and then an auto-ignition of hydrogen inside the tube may occur. To avoid the auto-ignition inside tube, it is necessary to understand the phenomenon with vortices generated near the contact surface.

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REFERENCES

1. Wolański, P., and Wójcicki, S., Investigation into the Mechanism of the Diffusion Ignition of a Combustible Gas Flowing into an Oxidizing Atmosphere, 14th Symp. (Int.) on Combustion, Pittsburgh, PA, The Combustion Inst., 1972, pp. 1217-1223.
2. Tanaka, K., Urano, Y., Iwasaka, M., and Hashiguchi, Y., Ignition at the Injection of High Pressure Hydrogen by a Rupture plate, *Koatsugasu*; **17**, No.7, 1978, pp. 333-338. (in Japanese)
3. Uejima, M., Watanabe, H., Noda, S., and Onuma, Y., Direct Numerical Simulation on the Spontaneous Ignition of Hydrogen Gas Jets, *Trans. Japan Soc. Mechanical Engineers B*, **64**, No. 626, 1998, pp. 3457-3463 (in Japanese)
4. Uejima, M., Watanabe, H., Noda, S., and Onuma, Y., Direct Numerical Simulation on the Spontaneous Ignition of Hydrogen Gas Jets (Further report), *Trans. Japan Soc. Mechanical Engineers B*, **69**, No.681, 2003, pp. 1269-1275 (in Japanese)
5. Liu, Y.F., Sato, H., Tsuboi, N., Higashino, F., and Hayashi, A.K., Numerical simulation on hydrogen fuel jetting from high pressure tank, *Sci. Tech. Energetic Materials*, **67**, 1, 2006, pp. 7-11.
6. Bazhenova, T.V., Bragin, M.V., Golub, V.V., Scherbak, S.B., and Volodin, V.V., Self ignition of the impulse hydrogen sonic jet emerging in the air semiconfined space, 25th International Symposium on Shock Waves, 2005, Paper 1118_1a.
7. Dryer, F.L., Chaos, M., Zhao, Z., Stein, J.N., Alpert, J.Y., and Homer, C.J., Spontaneous Ignition of Pressurized Releases of Hydrogen and Natural Gas into Air, *Combust. Sci. Technol.*, **179**, 2007, pp. 663-694.
8. Golub, V.V., Baklanov, D.I., Golovastov, S.V., Ivanov, M.F., Laskin, I.N., Saveliev, A.S., Semin, N.V., and Volodin, V.V., Mechanisms of High-pressure Hydrogen Gas Self-ignition in Tubes, *J. Loss Prevent. Process Ind.*, **21**, No. 2, 2008, pp. 185-198.
9. Mogi, T., Kim, D., Shiina, H., and Horiguchi, S., Self-ignition and Explosion During Discharge of High-pressure Hydrogen, *J. Loss Prevent. Process Ind.*, **21**, No. 2, 2008, pp. 199-204.
10. Yamada, E., Watanabe, S., Hayashi, A.K., and Tsuboi, N., Numerical Analysis on Auto-ignition of a High Pressure Hydrogen Jet Spouting from a Tube, *Proc. 32nd Int. Comb. Symp.*, **32**, 2009, pp. 2363-2369.
11. Xu, B.P., El Hima, L., Wen, J.X., Dembele, S., Tam, V.H.Y., and Donchev, T., Numerical Study on the Spontaneous Ignition of Pressurized Hydrogen Release through a Tube into Air, *J. Loss Prevent. Process Ind.*, **21**, No. 2, 2008, pp. 205-213.
12. Petersen, E.L., and Hanson, R.K., Reduced Kinetics Mechanisms for Ram Accelerator Combustion, *J. Propul. Power*, **15**, No. 4, 1999, pp. 591–600.
13. Chapman, S., and Cowling, T.G., The Mathematical Theory of Non-Uniform Gases, Cambridge University Press, UK, 1970, pp. 226-279.
14. Wilke, C.R., A Viscosity Equation for Gas Mixtures, *J. Chem. Phys.*, **18**, No. 4, 1950, pp. 517-519.

15. Cheng, T.S., Wehrmeyer, J.A., and Pitz, R.W., *Combust. Flame*, **91**, No.3-4, 1992, pp. 323-345.
16. Mizobuchi, Y., Tachibana, S., Shinjo, J., Ogawa, S., and Takeno, T., *Proc. 29th Int. Comb. Symp.*, **29**, 2002, pp.2009-2015.
17. http://www.paraview.org/