FUNDAMENTALS OF HYDROGEN IGNITION
and
HIGH PRESSURE HYDROGEN JET AUTO-IGNITION

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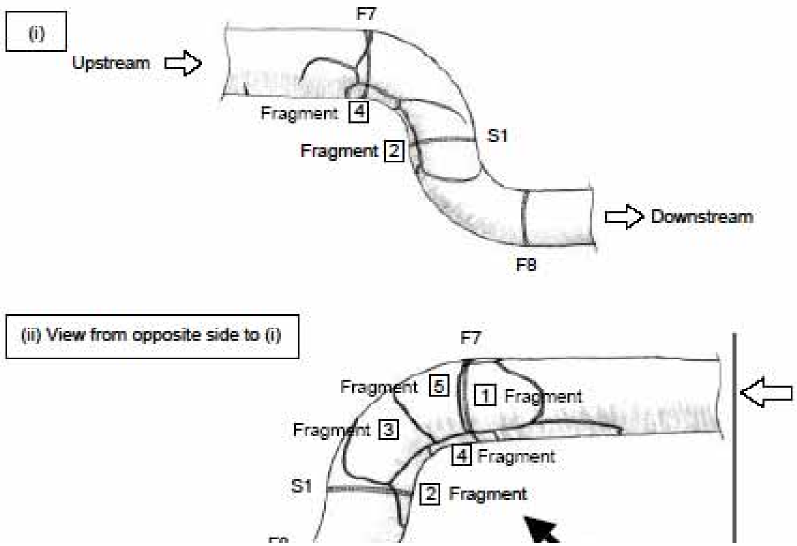
High Pressure Hydrogen Leak Issues

- Old story and still new: Chemical factory, refinery, etc. have hydrogen ignition caused by leaking. → In many cases ignition occurs when it leaks from a tube.
- New story: Fuel cell system will use a high pressure hydrogen. By accident hydrogen may leak and auto-ignite. We need a regulation for high pressure hydrogen uses.
- We still do not know in detail in which case hydrogen auto-ignites.

High Pressure Hydrogen Combustion Example

Hamaoka Reactor Accident Report



http://www.nisa.meti.go.jp/english/0207eng.pdf#search='hamaoka accident'

- **High pressure hydrogen was mixed with air and detonates. The original mixture pressure was assumed to be 5-8 MPa and the pipe received a combustion pressure (most probably detonation) of 180-300 MPa.**

Fundamentals of Hydrogen Ignition

The First Part of Lectures

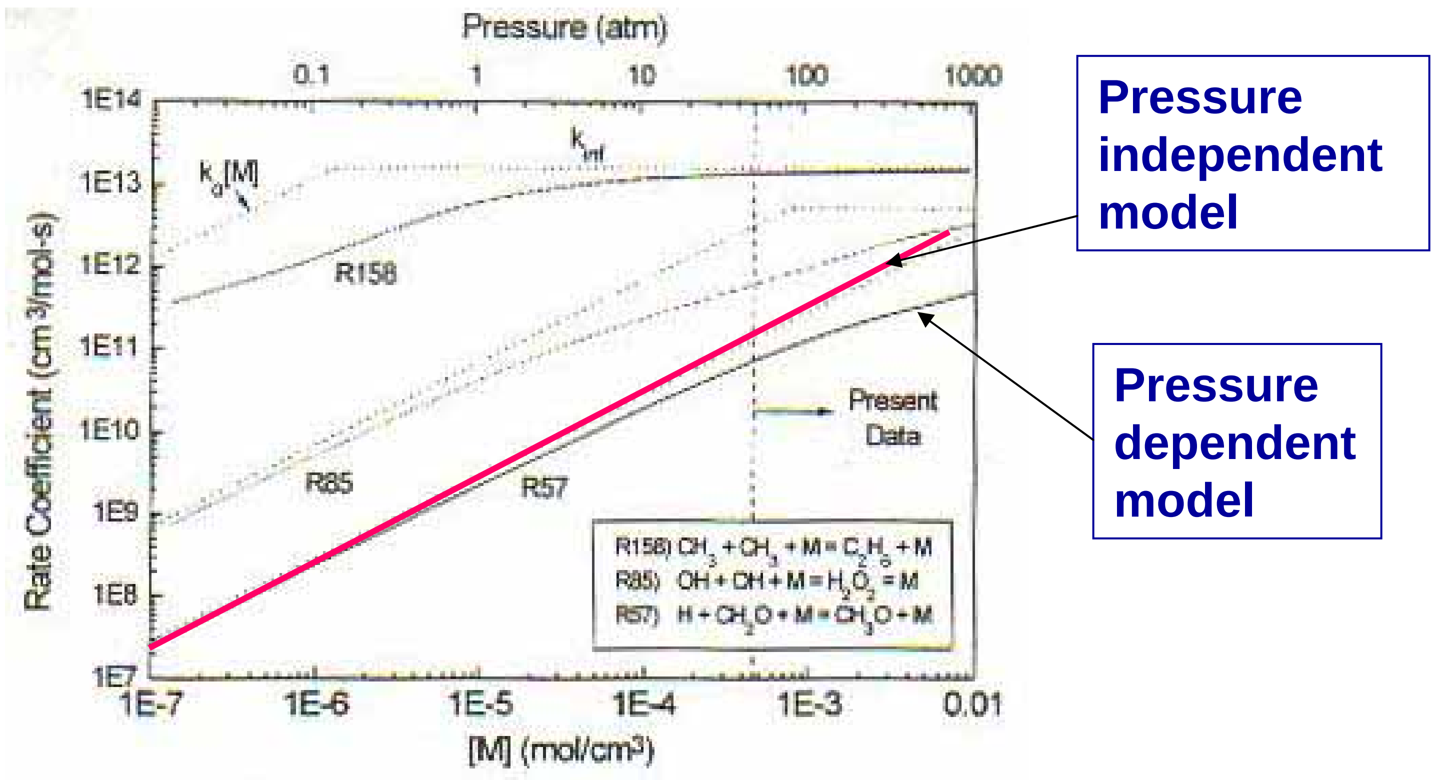
Why the hydrogen ignition fundamentals?

↓

Because we use the hydrogen data for low pressure case in most cases.

This part of my lecture will show you how much the low pressure chemistry does not agree with the high pressure chemistry using a detonation calculation.

Fundamentals of Hydrogen Ignition (1)
Present Status of Hydrogen Combustion Model and Its Issues



Fundamentals of Hydrogen Ignition (2)

Numerical Analysis

- 1D compressible Euler equations
- Fuel →hydrogen,   Oxidizer →oxygen or air
- The convective terms: 2nd order Harten-Yee type TVD scheme
- The unsteady terms: Strang type fractional step method
- Axisymmetric flow
- Chemical reactions : Petersen & Hanson model, Koshi model
- Computational grids: decided by having 33 points in a half reaction length
- Initial pressures: 0.1, 0.2, 0.5, 1, 2, 3, 4, 5, 6, 7 ,8, 9, 10 MPa
- Initial temperature: 298.15 K
- The computer used: NEC SX-6 (Max 8CPU) at JAXA/ISAS

Fundamentals of Hydrogen Ignition (3)

-Effects of initial pressure on detonation initiation-

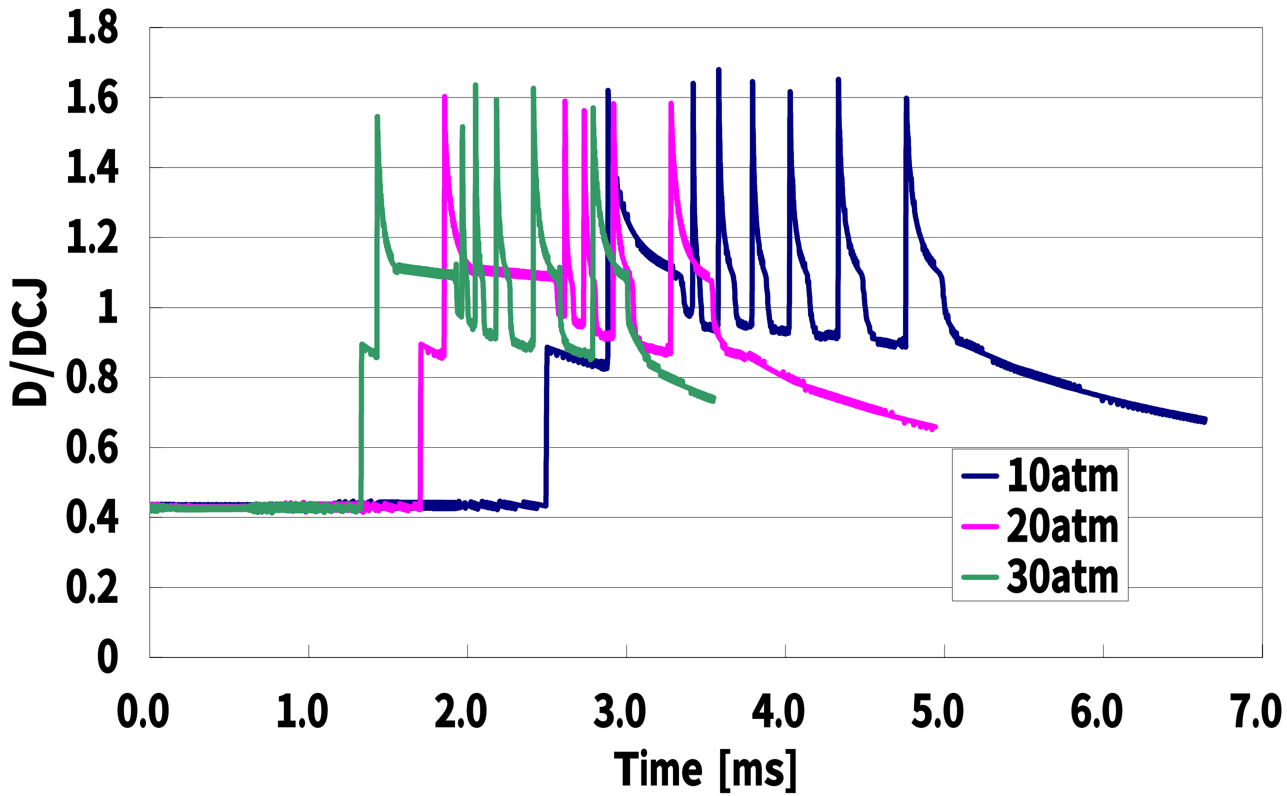
Initial pressure P_0[MPa]	Grid size [μ m]	Ignition pressure Pi[times]	Ignition temperature Ti[K]	Ignition area Ai [points]
0.1	1.71	38	1400	1000
0.2	0.51	38	1400	4500
0.5	0.275	38	1600	3000
1.0	0.16	20	1400	9000
2.0	0.12	20	1400	9000
3.0	0.09	20	1400	9000
4.0	0.12	40	2000	1000
5.0	0.06	40	2000	3000
6.0	0.05	40	2000	4500
7.0	0.044	40	2000	3000
8.0	0.038	40	2000	1000
9.0	0.032	40	2000	1000
10.0	0.03	40	2000	3000

•Ignition pressure is very high for the case of the initial pressures higher than 5 MPa probably due to near supercritical conditions.

•The cases of the initial pressures; 1.o, 2.0, and 3.0 MPa, need less ignition pressure due to high sensitivity for such cases.

Fundamentals of Hydrogen Ignition (4)

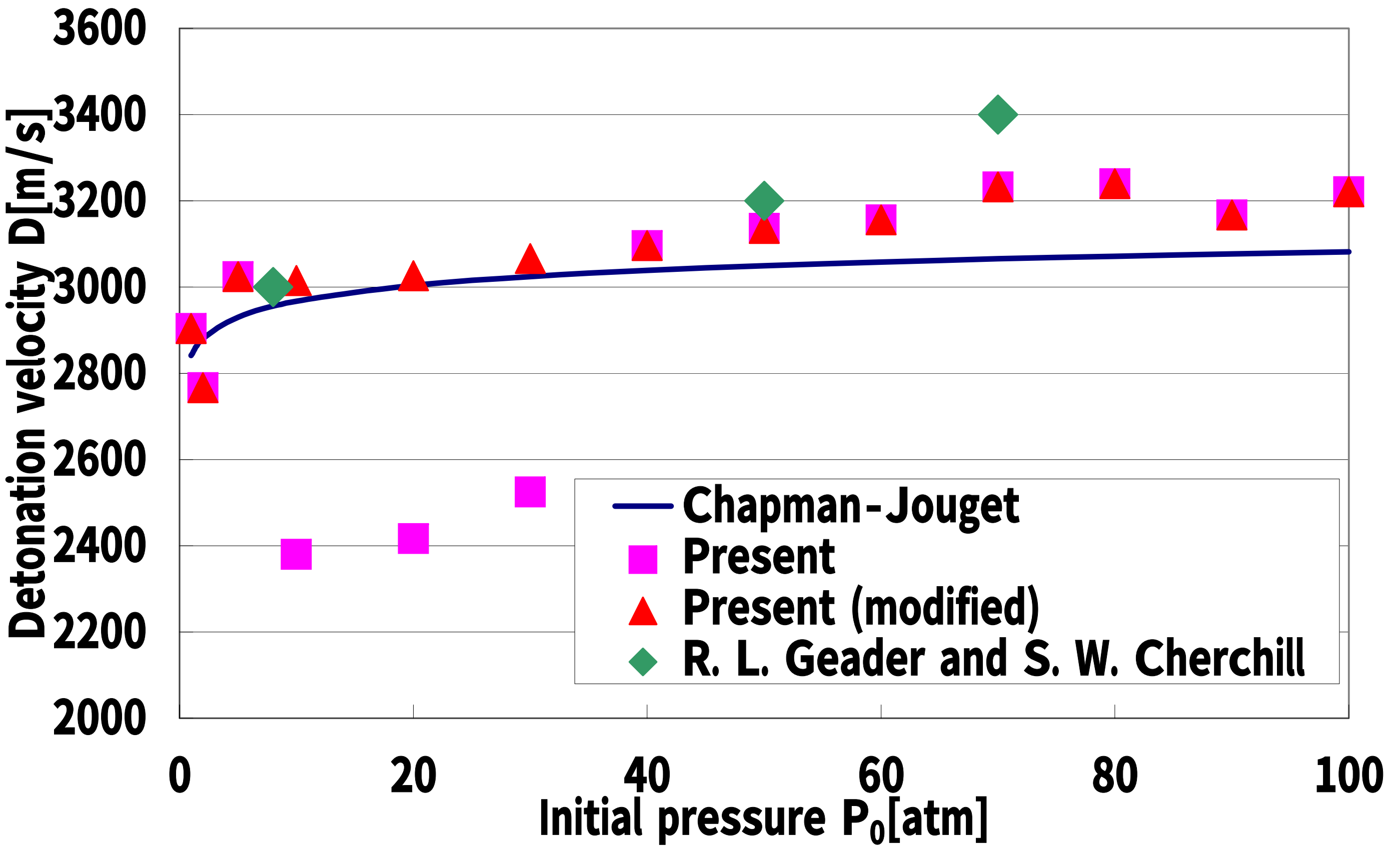
-Effects of initial pressure on detonation initiation-



- Detonation velocity profiles for the cases of the initial pressures of 1.0, 2.0, 3.0 MPa. 1D calculation of detonation gives typically such velocity profile.

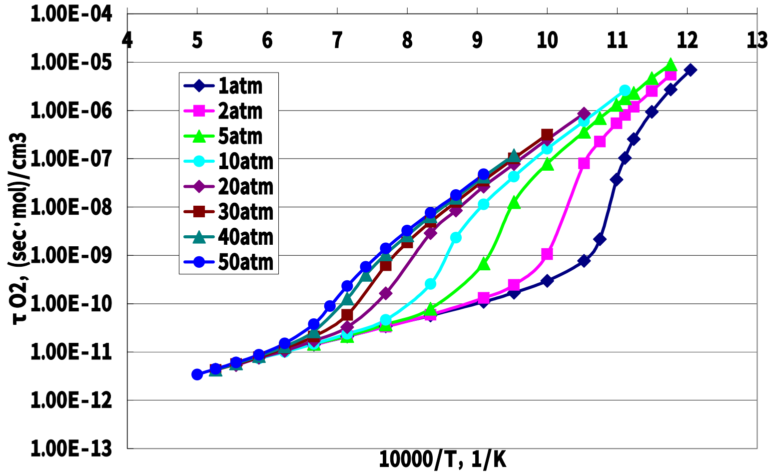
Fundamentals of Hydrogen Ignition (5)

-Effects of initial pressure on detonation initiation-

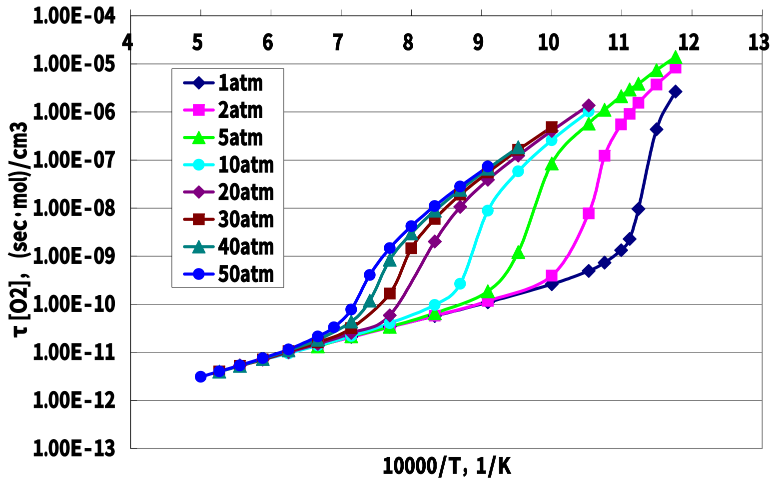


•The “present” includes an expansion region and damping region of velocity for average calculation and “present (modified) does not an expansion region.

Fundamentals of Hydrogen Ignition (6)
-High pressure chemical reaction model (1)-



Petersen & Hanson model

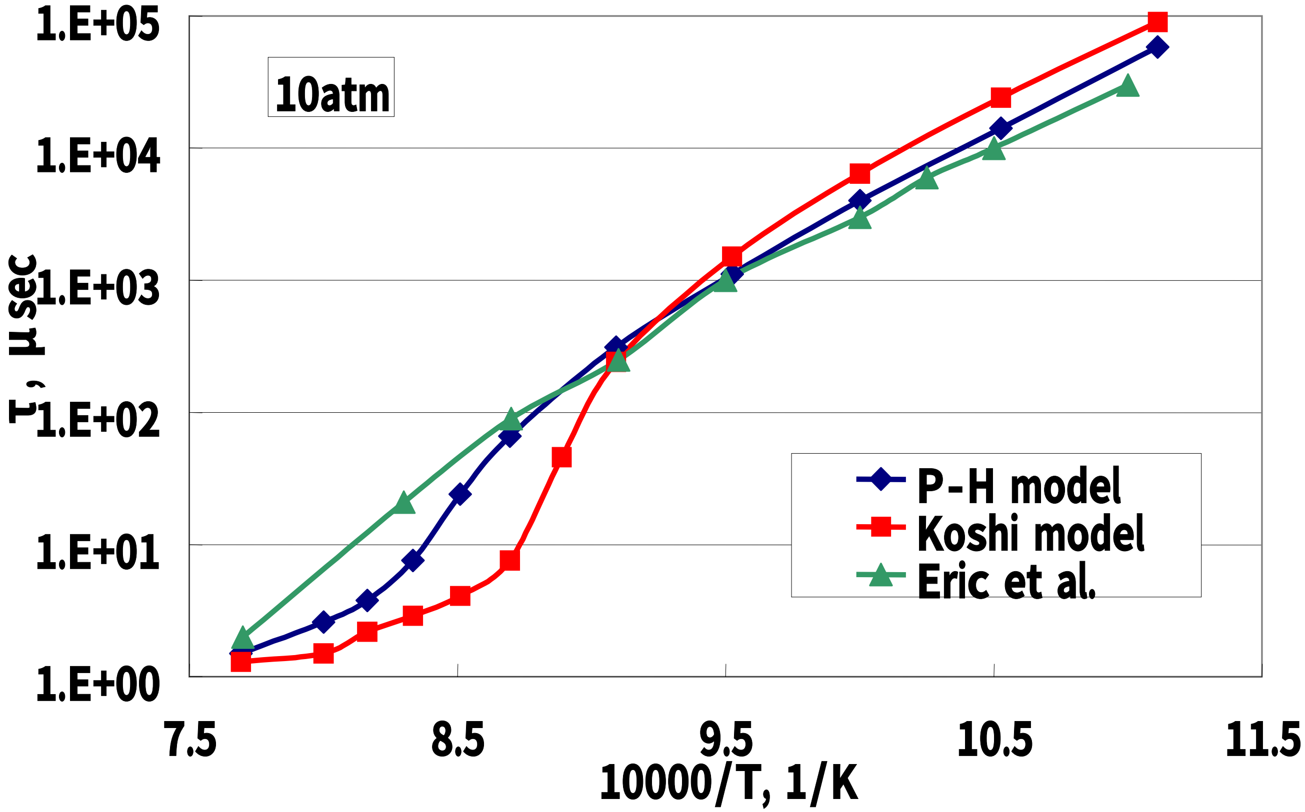


Koshi model

•Ignition delay time using CHEMKIN 4.1: in these results both model gives good results and it is difficult to see the difference in the results.

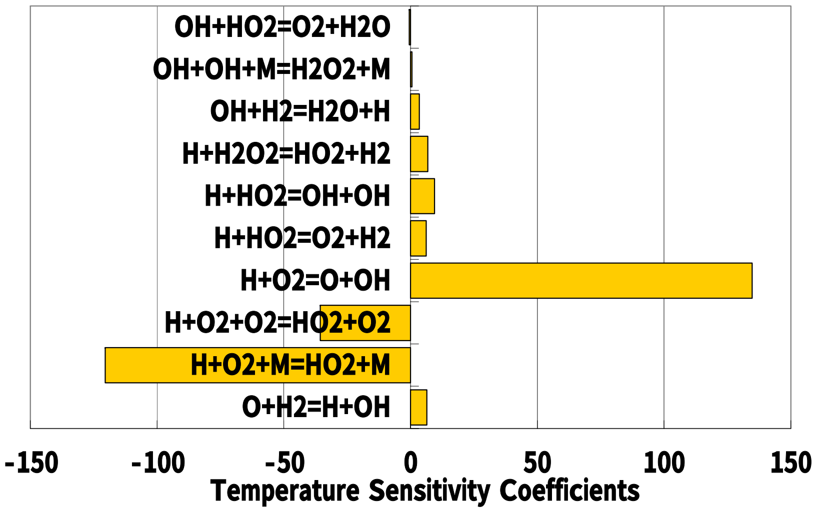
Fundamentals of Hydrogen Ignition (7)

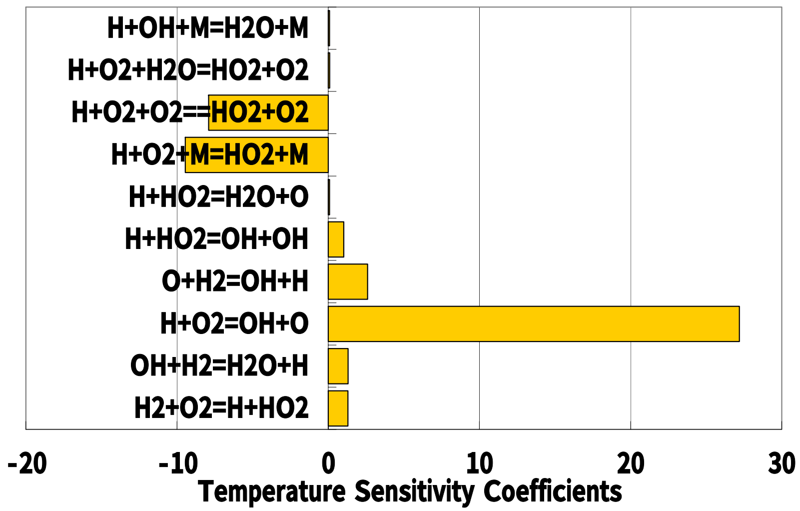
-High pressure chemical reaction model (2)-



- Koshi model gives shorter delay time than P-H model at higher temperature.

Fundamentals of Hydrogen Ignition (8)
-High pressure chemical reaction model (3)-



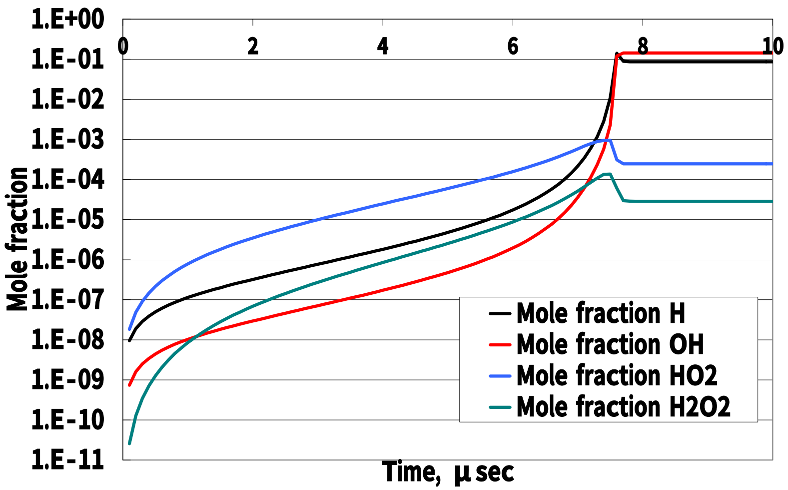


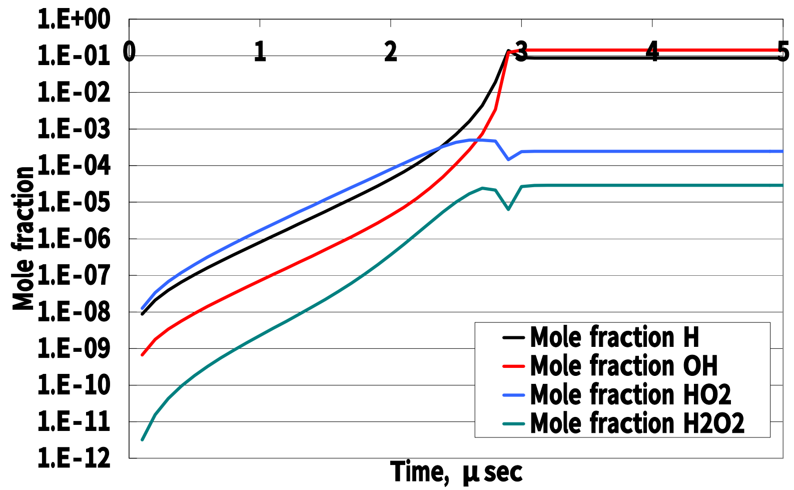
Petersen & Hanson model

Koshi model

•P-H model shows H+O2+M->HO2+M sensitivity is much larger than that of Koshi model. Koshi model produces more radicals than P-H model.

Fundamentals of Hydrogen Ignition (9)
-High pressure chemical reaction model (4)-





Petersen & Hanson model

Koshi model

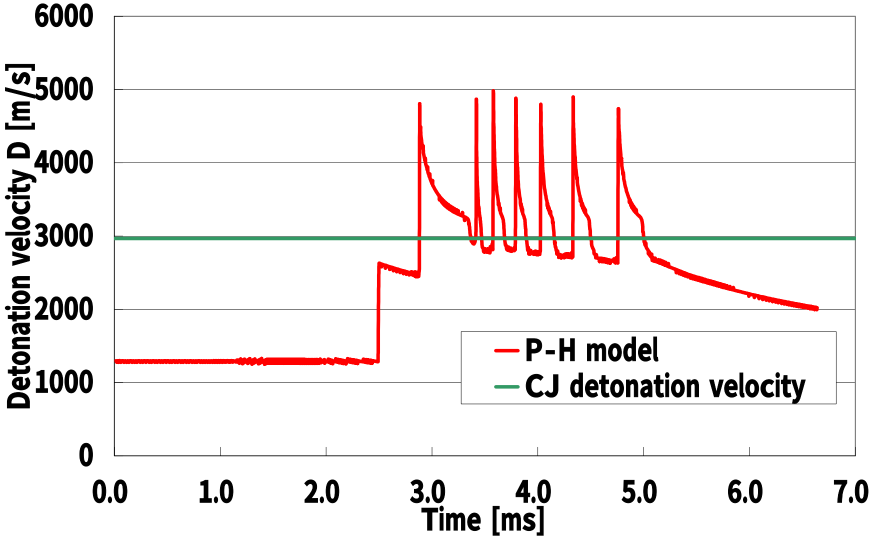
- Koshi model produces more radicals than P-H model.

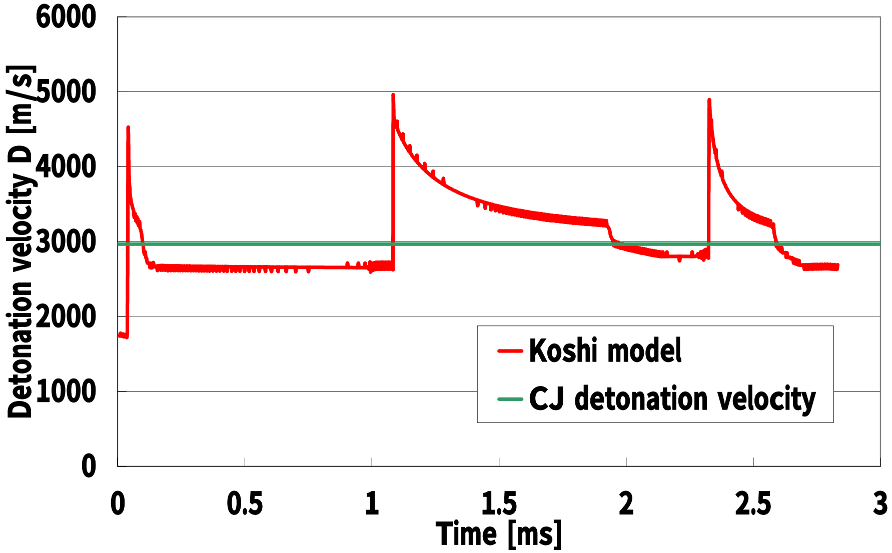
Fundamentals of Hydrogen Ignition (10)
-High pressure chemical reaction model (5)-

Reaction model	Grid size [μ m]	Ignition pressure P_i [times]	Ignition temperature T_i [K]	Ignition area Ia [points]
P-H model	0.16	20	1400	9000
Koshi model	0.16	50	2000	9000

- Koshi model provides a stable calculation, while P-H model does not.

Fundamentals of Hydrogen Ignition (11)
-High pressure chemical reaction model (6)-





Petersen & Hanson model

Koshi model

- **Koshi model gives a stable detonation velocity profiles, while P-H model does less stable ones..**

Fundamentals of Hydrogen Ignition (12)

Summary

- The pressure dependent chemical reaction model is important to solve the high pressure hydrogen auto-ignition problems.
- This part shows that we needs such pressure dependent reaction mechanism provides the better explanation of experimental data at high pressure atmosphere.

High Pressure Hydrogen Jet Auto-Ignition

The Second Part of Lectures

Background

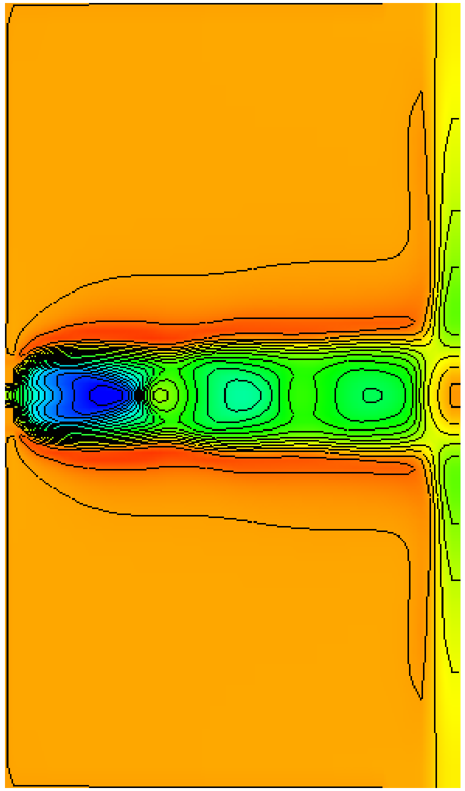
Experimental findings

- **Wolanski & Wojicicki (1973): High pressure H_2 jet auto-gnition was found.**
- **Hayashi et al. (2004): High pressure He jet hitting a wall.**
- **Bazhenova et al. (2005): Similar High Pressure H_2 jet from a hole.**
- **Dryer et al. (2006): High pressure H_2 jet auto-ignition from a tube.**
- **Mogi et al. (2006): High pressure H_2 jet auto-ignition from a hole.**
- **Pinto et al. (2007) & Aizawa et al. (2007): High pressure H_2 jet auto-ignition from a hole and a tube.**
- **Mogi et al. (2008): High pressure H_2 jet auto-ignition from a tube.**

Numerical findings

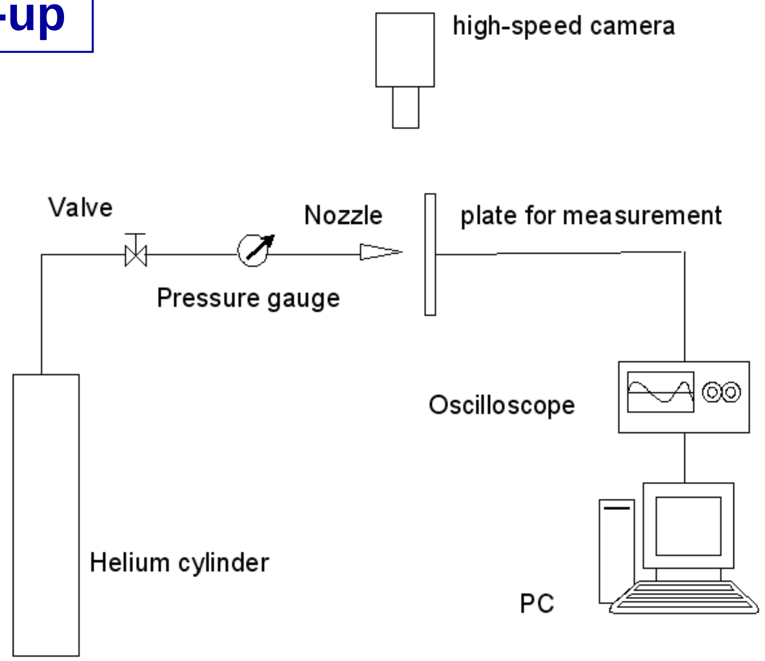
- **Hayashi et al. (2004): High pressure He jet hitting a wall.**
- **Liu et al. (2005): High pressure H_2 jet leak from a hole.**
- **Yamada et al. (2008): High pressure H_2 jet leak from a tube.**
- **Xu et al. (2008): High pressure H_2 jet released from a tube.**

High Pressure Hydrogen jetted to an obstacle



High Pressure Hydrogen jetted to an obstacle(1)

Experimental set-up



Reservoir tank pressure [MPa]	Reservoir tank temperature [K]	Distance between the nozzle exit and the plate [mm]	Nozzle exit initial pressure [MPa]	Nozzle exit initial temperature [K]
100	300	10, 30, 50	0.1 (air)	300 [K]

High Pressure Hydrogen jetted to an obstacle(2)

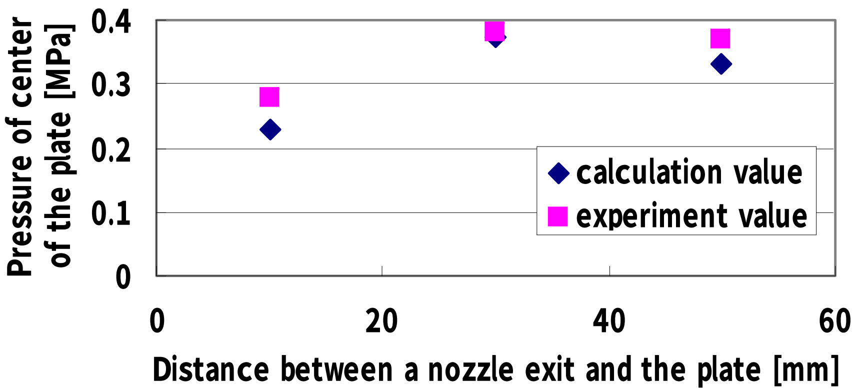
Numerical Analysis

- 2D compressible Euler equations
- Fuel →hydrogen,   Oxidizer →air
- The convective terms: Harten-Yee type TVD scheme
- The unsteady terms: Strang type fractional step method
- Axisymmetric flow
- No chemical reactions due to a use of He as a fuel
- No viscous and thermal conductivity effects
- Specific heats are a function of temperature.
- No bulk viscosity, no Soret effects, no Dufour effects, no pressure gradient diffusion, no gravity effects.
- The boundary condition at the wall is adiabatic and slip.

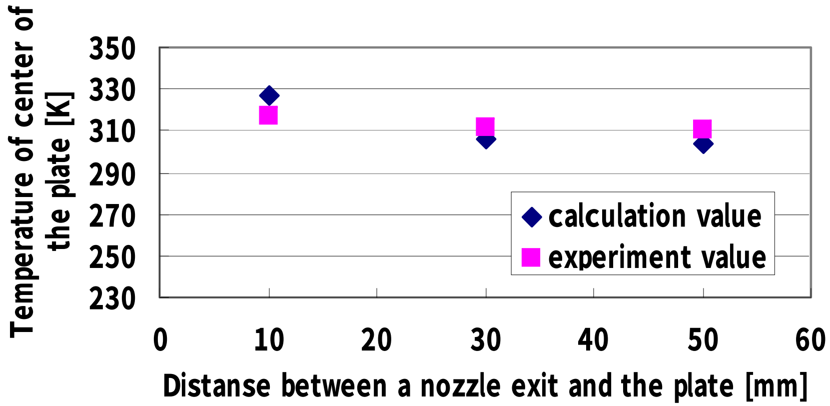
High Pressure Hydrogen jetted to an obstacle(3)

-Comparison between experimental and numerical results-

Pressure profiles



Temperature profiles

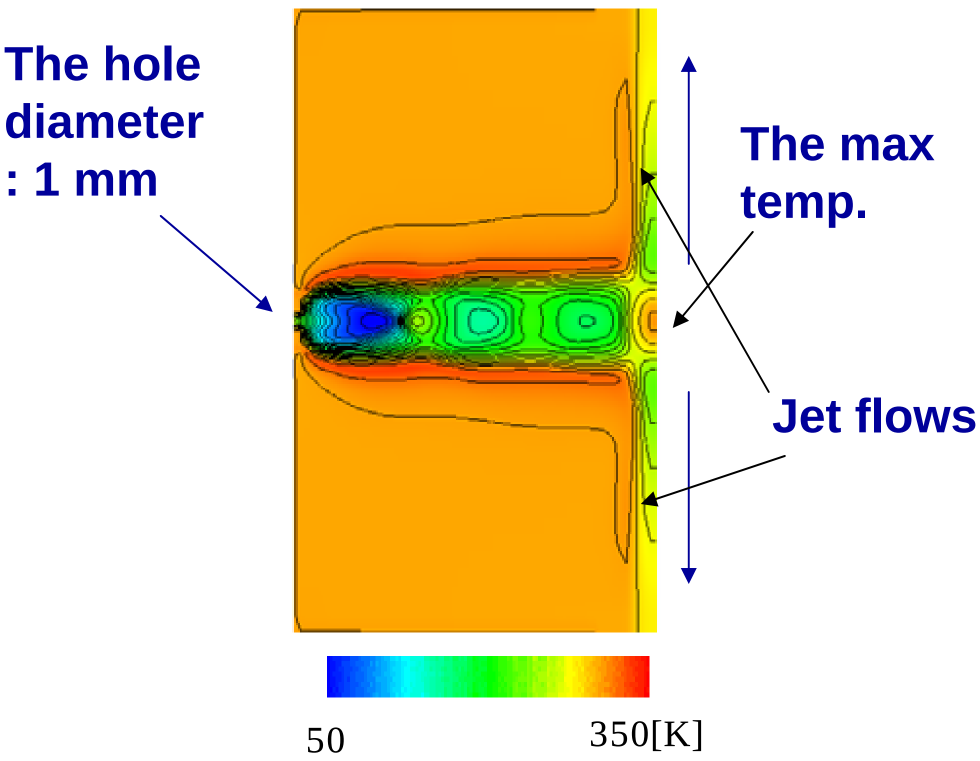


- **The slight difference comes from He (experiments) and H2 (computations).**
- **Temperature and pressure become constant being longer between the nozzle exit and the plate surface.**

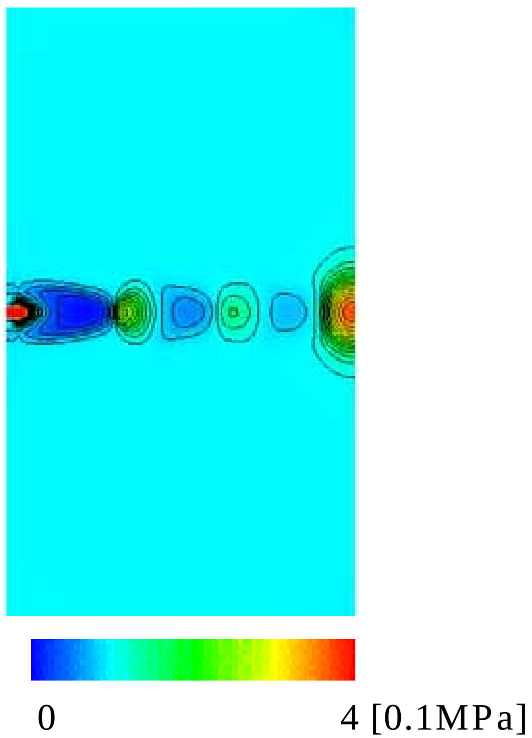
High Pressure Hydrogen jetted to an obstacle(4)

-Temperature and pressure profiles between the nozzle exit and the plate surface (Reservoir pressure at 10MPa)-

Temperature profiles



Pressure profiles

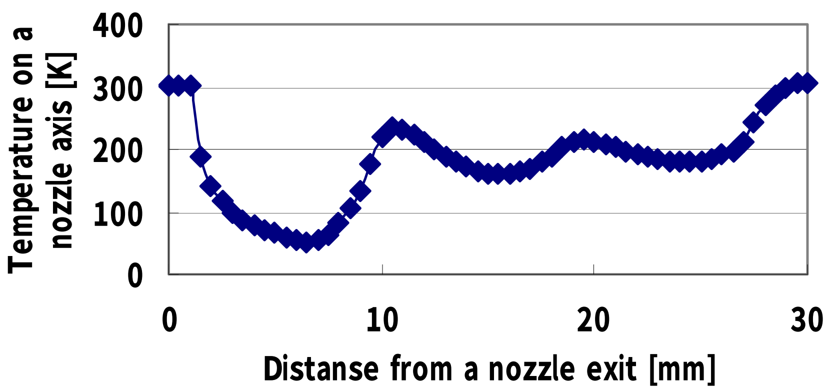


- The distance between the nozzle exir and the plate surface : 30 mm

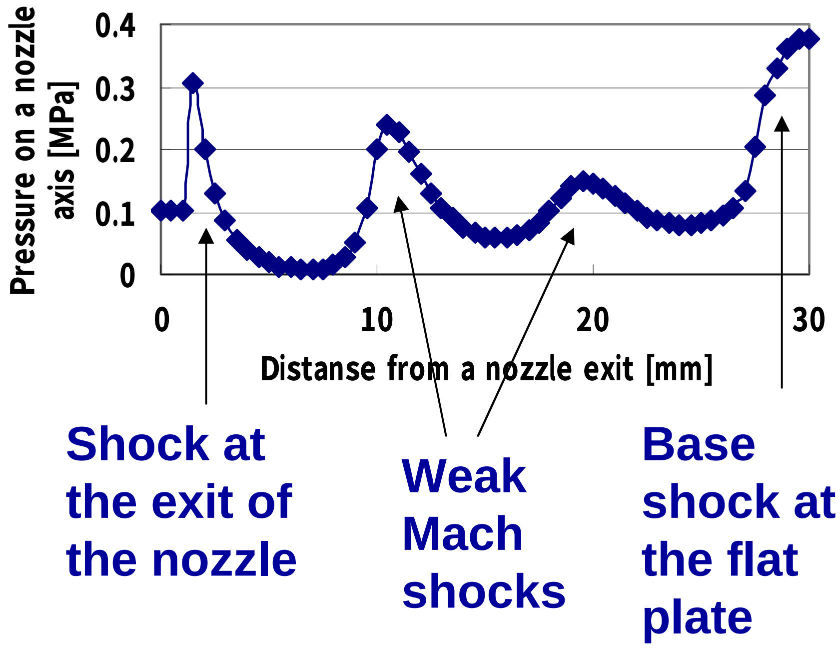
High Pressure Hydrogen jetted to an obstacle(5)

-Temperature and pressure profiles at the center axis of the jet between the nozzle exit and the plate surface (Reservoir pressure at 10MPa)-

Temperature profiles



Pressure profiles

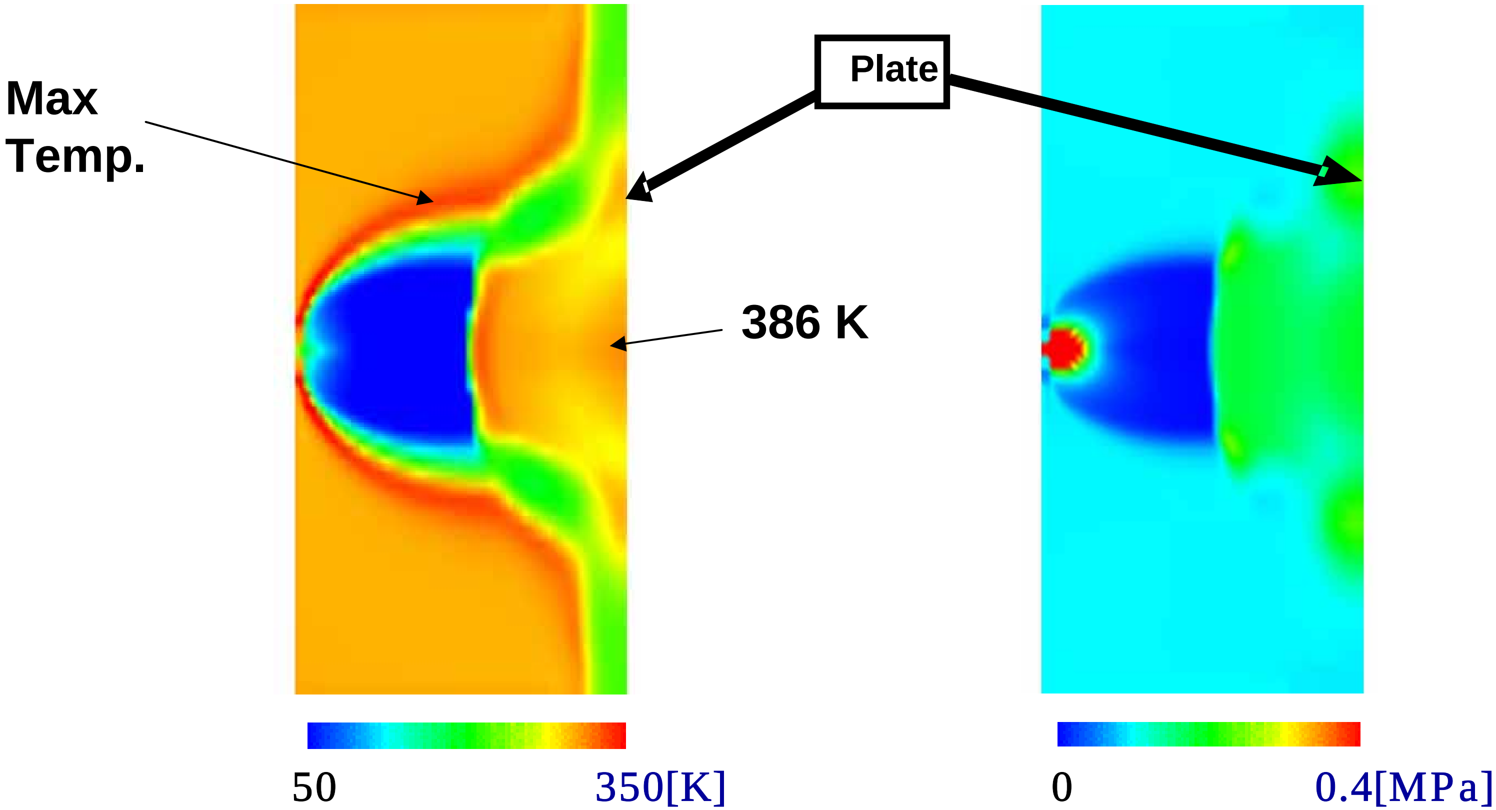


•The base shock is somewhat strong (0.38 MPa), but the temperature there is about 303 K.

High Pressure Hydrogen jetted to an obstacle(6)

-Temperature and pressure profiles between the nozzle exit and the plate surface (Reservoir pressure at 100MPa)-

Temperature profiles | Pressure profiles

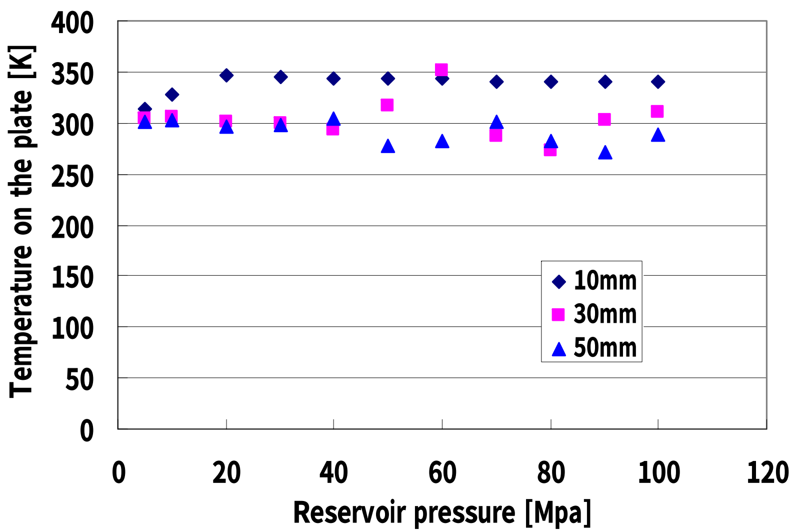


• The temperature at the separation bulb is 386 K not to ignite H2.

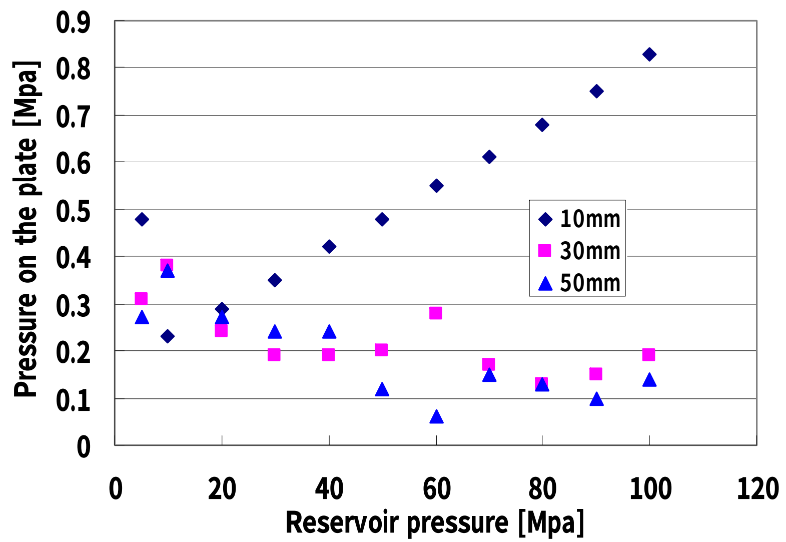
High Pressure Hydrogen jetted to an obstacle(7)

-Temperature and pressure profiles on the plate (Reservoir pressure at 100MPa)-

Temperature profiles



Pressure profiles

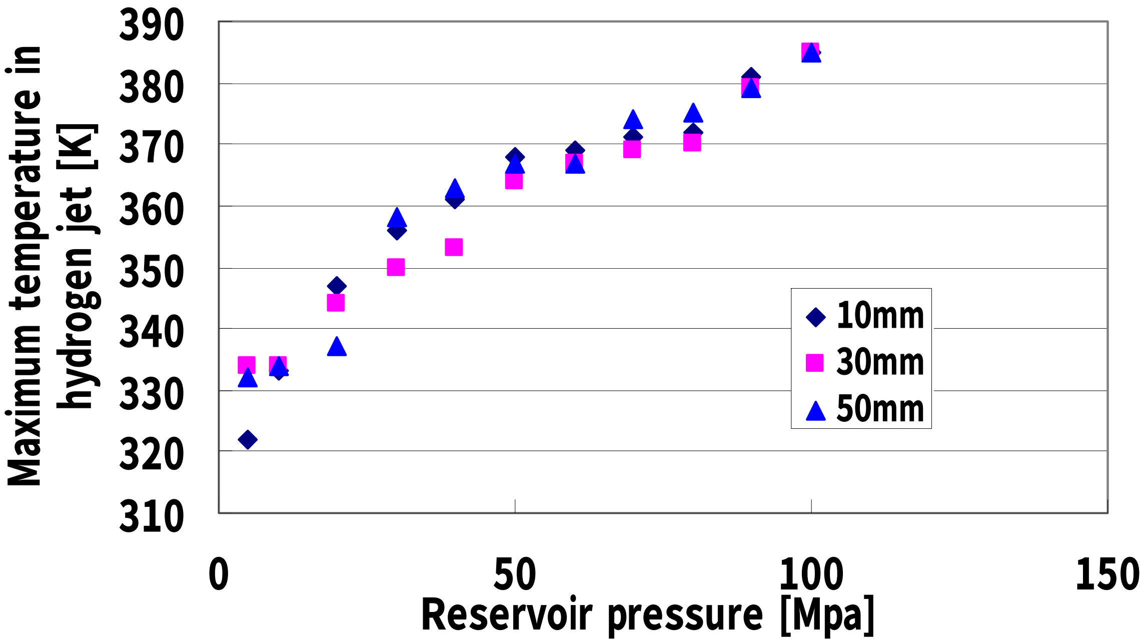


- Temperature profiles do not change much to reservoir pressure.
- But pressure profiles do change much especially for the short distance between the nozzle exit and the plate.

High Pressure Hydrogen jetted to an obstacle(8)

-Max_temperature profiles in the jet (Reservoir pressures from 5 to 100MPa)-

Max temperature profiles



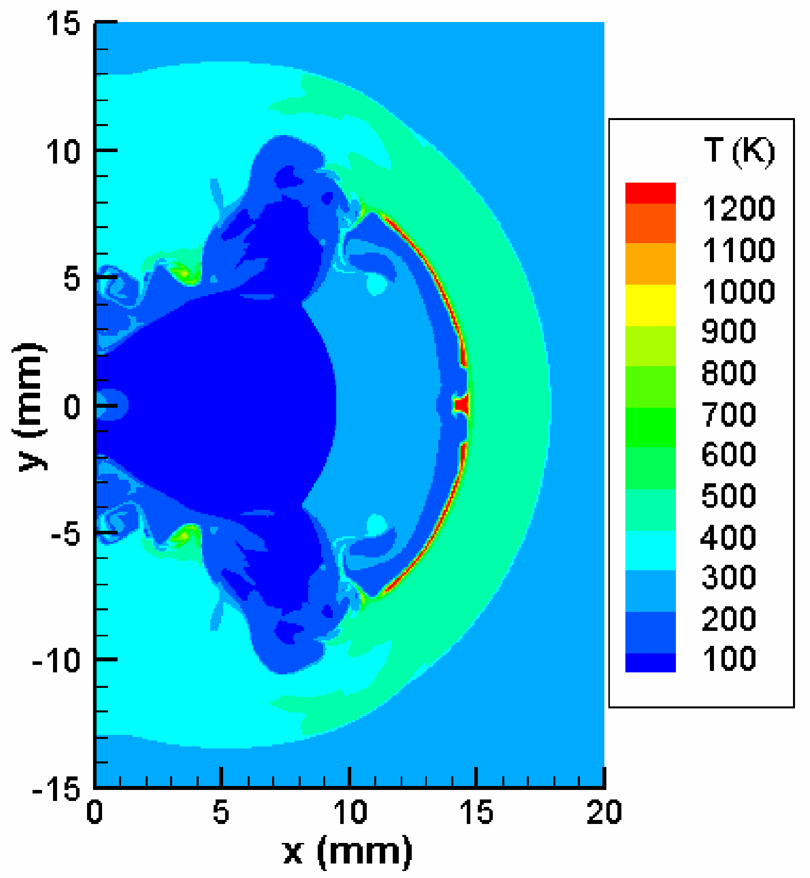
- Max temperature profiles increase as the reservoir pressure increase.
- But temperature is still lower than ignition temperature.

High Pressure Hydrogen jetted to an obstacle(9)

Summary

- The numerical temperature agrees well with the experimental one on the surface of the flat plate.
- The temperature profiles on the flat plate for the case of the reservoir pressure of 100 MPa are within 350 K, which does not give the ignition to hydrogen.
- Just only for the case of 10 mm distance between the nozzle exit and the flat plate surface, the maximum temperature shows a linear increase of temperature to the increase of reservoir pressure.

High Pressure Hydrogen Leak from a small Hole



High Pressure Hydrogen Leak from a small Hole(1)

Numerical Analysis

- 2D axisymmetric Euler equations with the detailed chemical reaction mechanism.
- Hydrogen reaction mechanism of 9 species and 18 reactions.
- The convection terms are integrated by Harten-Yee type non-MUSCL TVD scheme.
- The time discretization is performed by Strang-type fractional step method.
- Chemical reactions are treated by a point implicit way to avoid stiffness.

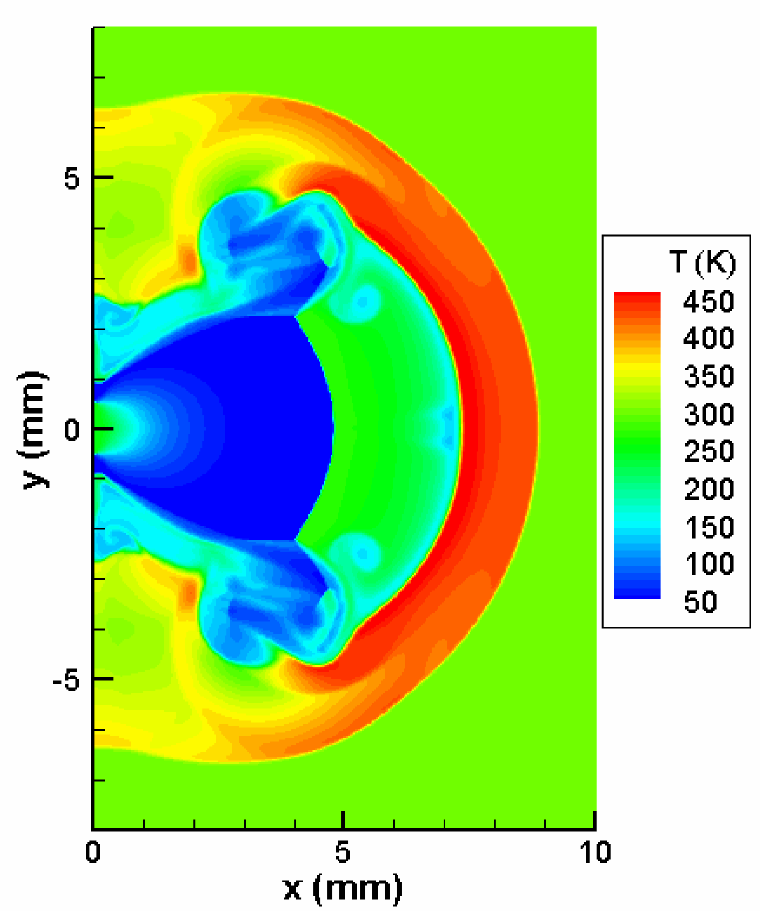
High Pressure Hydrogen Leak from a small Hole(2)

Numerical Conditions

- Reservoir tank pressures are 10, 40, 70 MPa.
- Reservoir tank temperature is 300 K.
- The computational domain is rectangular.
- The left boundary condition is a solid wall and other boundaries are free stream conditions.
- A small hole of 1, 3, 5 mm diameter is located at the center of the left wall.
- The H2 jet is choked at the hole and the Mach number there is unity.
- The initial ambient air is at 0.1 MPa, 300 K, and at rest.
- The uniform grid size of dx=dy=10 or 20 μm.

High Pressure Hydrogen Leak from a small Hole(3) -The case of the reservoir pressure of 10 MPa-

Temperature profiles

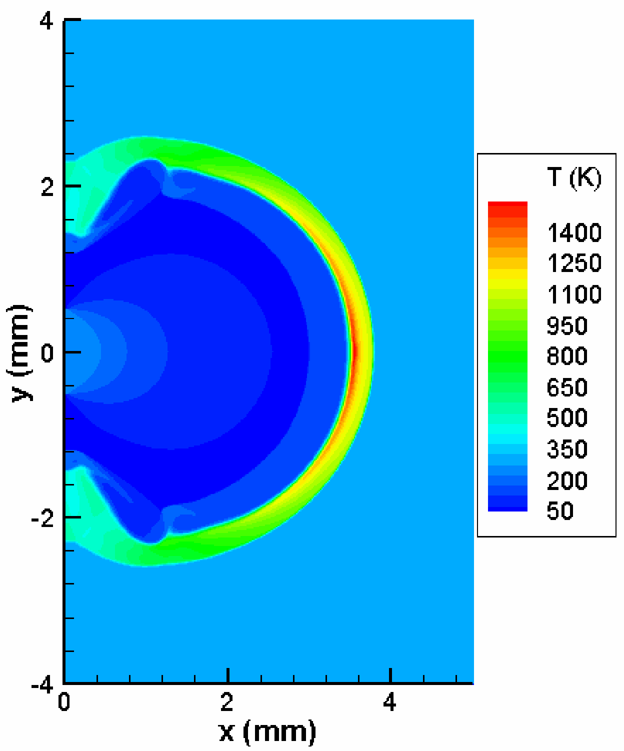


The time is at 10 μs.

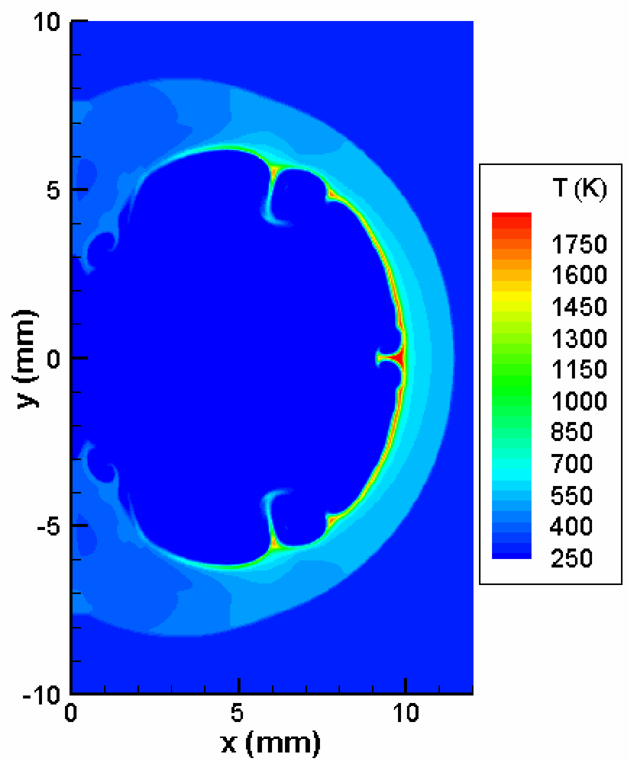
- **The max temperature is 450 K at the jet front. No OH molecules appear in this case.**

High Pressure Hydrogen Leak from a small Hole(4) -The case of the reservoir pressure of 40 MPa-

Temperature profiles



The time is at 2 μs.



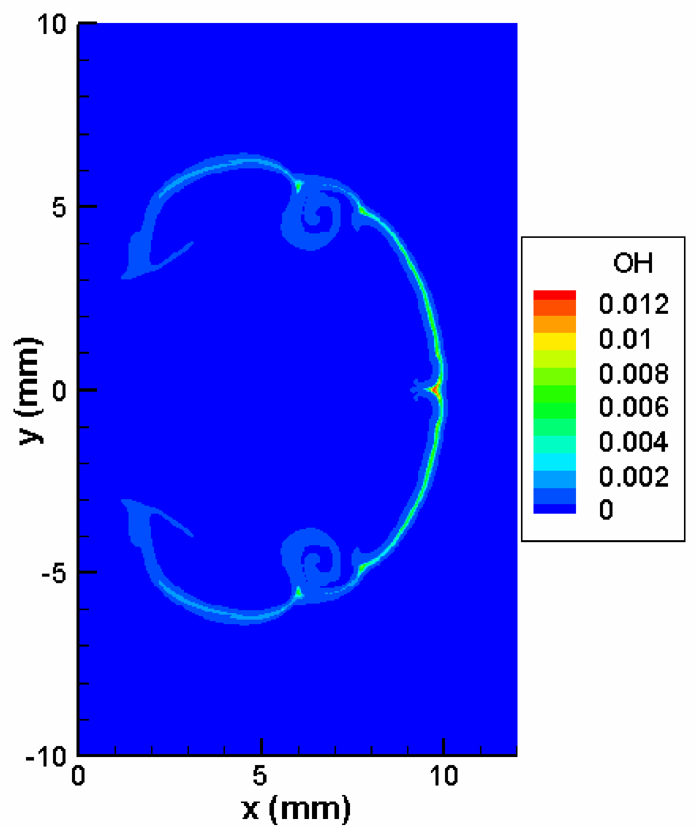
The time is at 10 μs.

- The max temperature is 1750 K at the contact surface (t=10 μs).

High Pressure Hydrogen Leak from a small Hole(5) -The case of the reservoir pressure of 40 MPa-

OH molecule profiles

The time is at 10 μs.

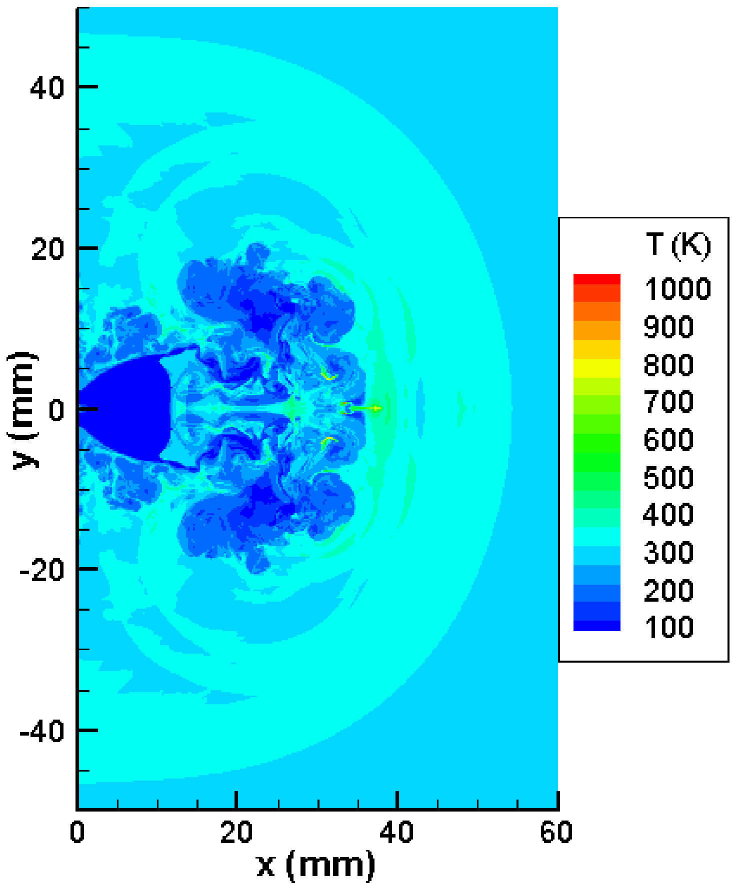


- A large number of OH molecules are found at the contact surface region.

High Pressure Hydrogen Leak from a small Hole(6)
-The case of the reservoir pressure of 40 MPa-

Temperature profiles

The time is at 100 μs.

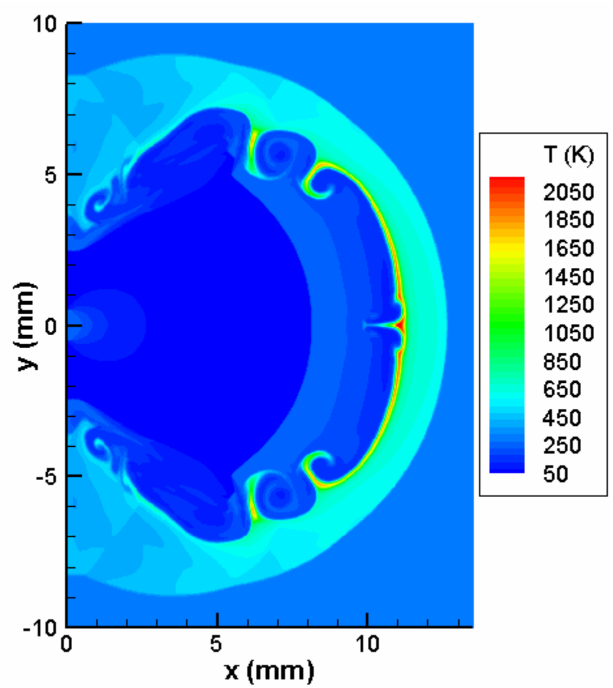


- When the jet propagates further, temperature goes down to extinguish the local flame.

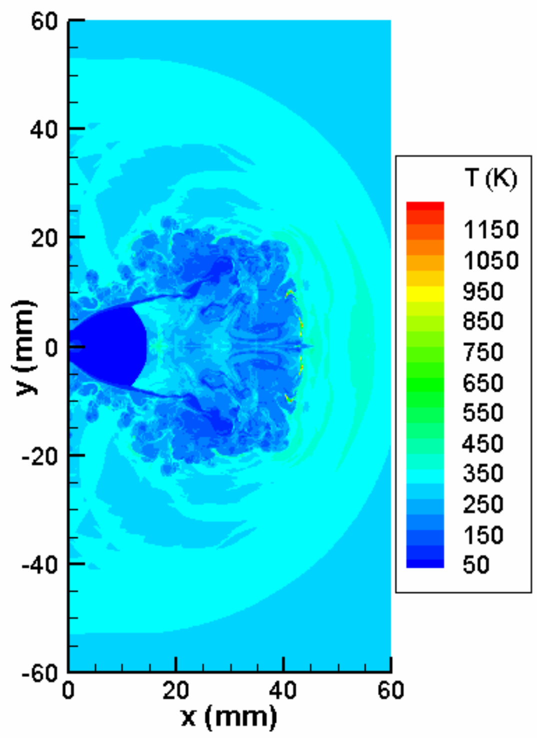
High Pressure Hydrogen Leak from a small Hole(7) -The case of the reservoir pressure of 70 MPa-

Temperature profiles

The time is at 10 μs.



The time is at 100 μs.

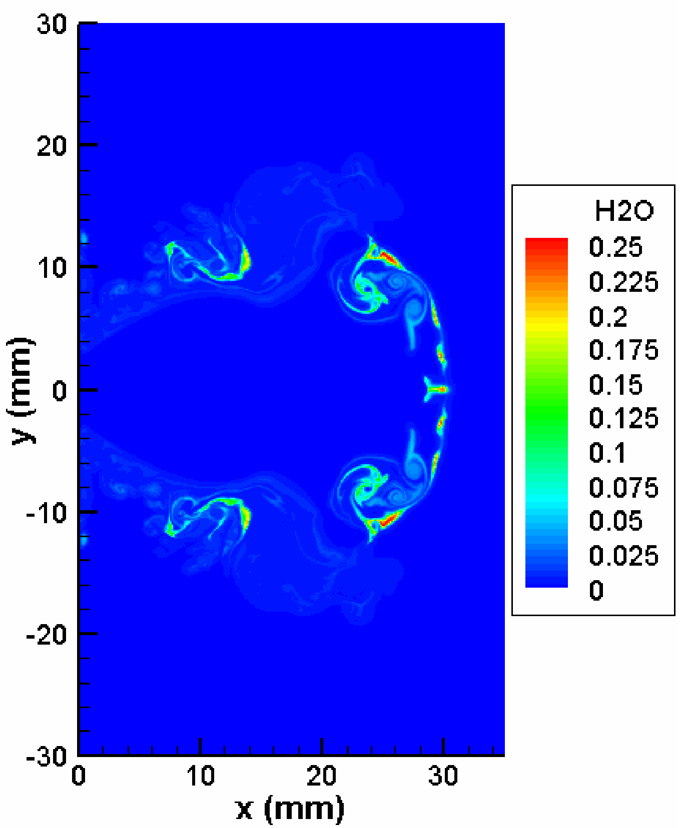


- **The over-expansion situation gives such quenching phenomena to the jet flame.**

High Pressure Hydrogen Leak from a small Hole(8)
-The case of the reservoir pressure of 70 MPa-

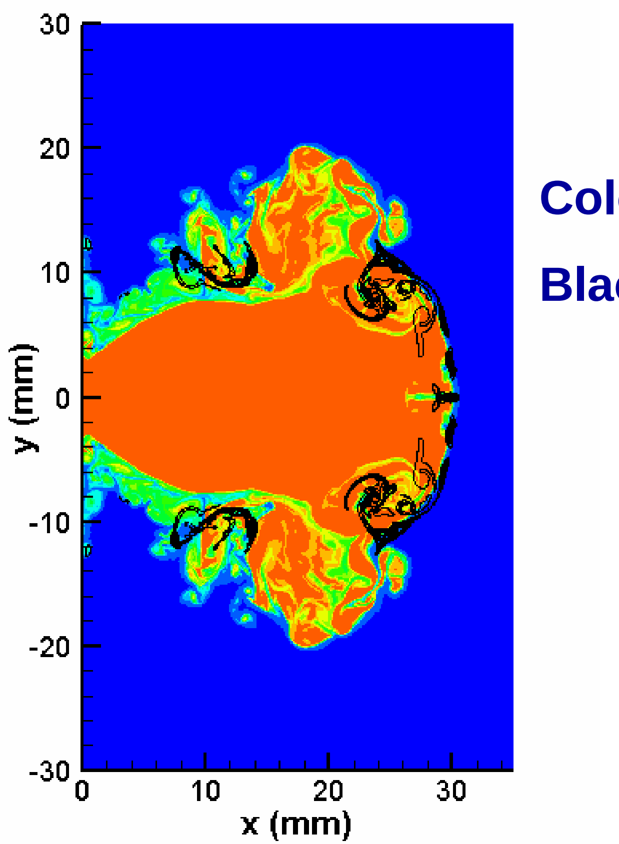
H2O mass fraction profiles

The time is at 60 μs.



H2O and H2 mass fraction profiles

The time is at 60 μs.



Color: H2

Black: H2O

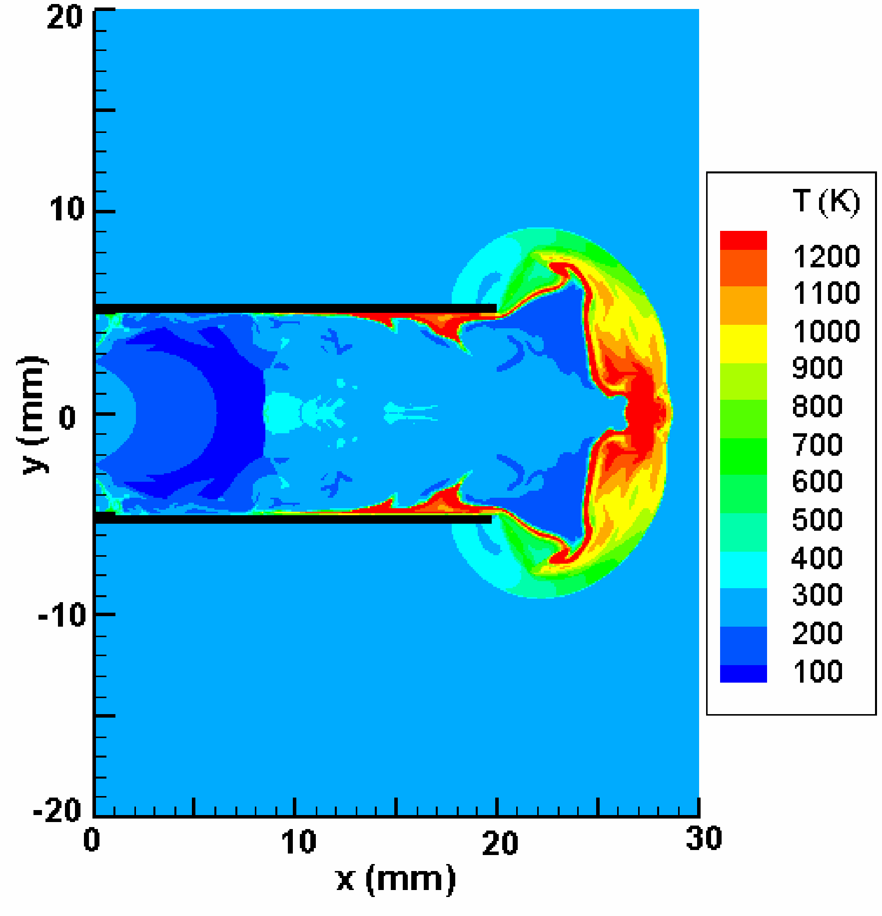
- **H2O stays between hydrogen and air and propagates together with the jet. H2O prevents H2 and air mixing.**

High Pressure Hydrogen Leak from a small Hole (9)

Summary

- For 40 and 70 MPa cases the local combustion occurs at the early stage of H2 jet propagation. The high temperature region behind the leading shock wave provides such local ignition.
- In all cases the max temperature decreases below 1000 K at t=100 μs. H2O prevents the well mixing of H2 and air.
- The local combustion does not stay and moves together with the jet not to keep such ignition source.

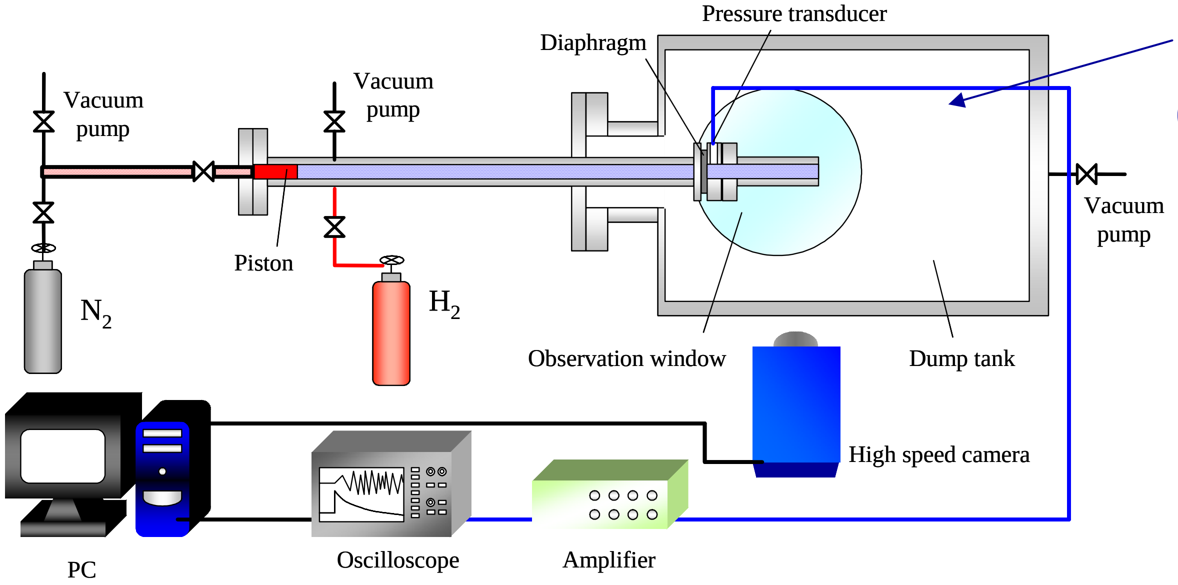
High Pressure Hydrogen Leak from a Tube



High Pressure Hydrogen Leak from a Tube(1)

Experimental set-up

Small shock tube system



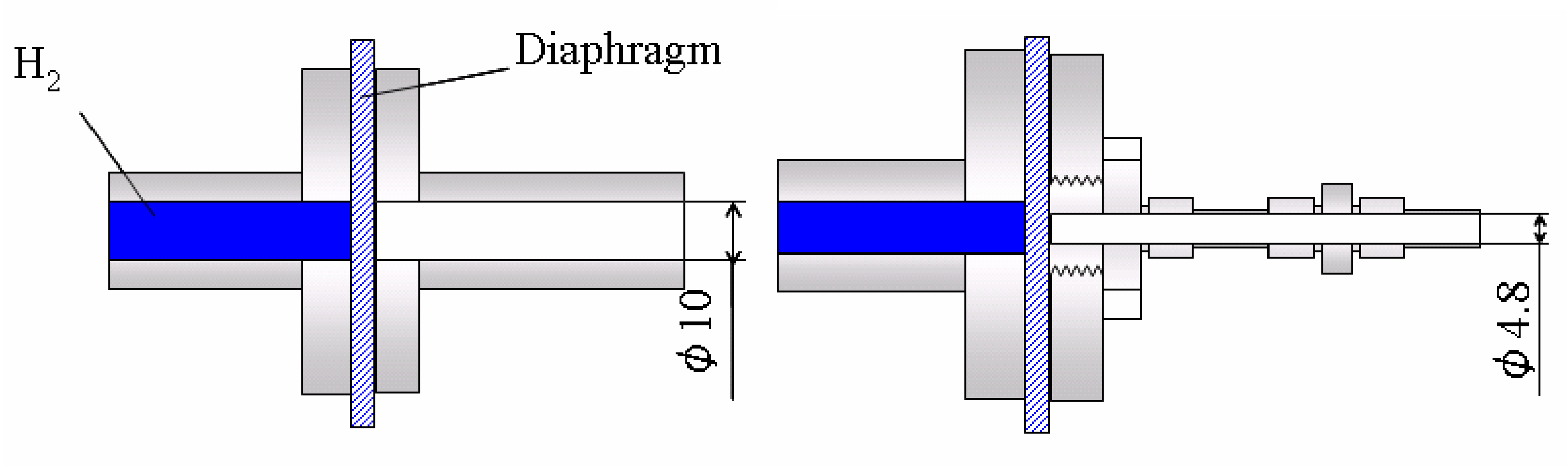
Dump tank pressure= 0.1 MPa

High pressure section (N2) P_H [MPa], T_H [K]	Low Pressure section (H2) P_L [MPa], T_L [K]	Hydrogen jet pressure [MPa]	Tube 1 d_i inner diameter L tube length [mm]	Tube 2 d_i inner diameter L tube length [mm]
8 , 300	0.9, 300	3.8, 6.8	d_i=4.8, L=48, 71, 113	d_i=10.0 L=50, 100, 180

High Pressure Hydrogen Leak from a Tube(2)

Tube configuration

Diaphragm: 50 μm thickness



Tube 1 d_i inner diameter [mm] L tube length [mm]	Tube 2 d_i inner diameter [mm] L tube length [mm]
d_i=4.8, L=48, 71, 113	di=10.0 L=50, 100, 180

High Pressure Hydrogen Leak from a Tube (3)

Numerical Analysis

- 2D axisymmetric Euler equations with the detailed chemical reaction mechanism.
- Hydrogen reaction mechanism of 9 species and 18 reactions.
- The convection terms are integrated by Harten-Yee type non-MUSCL TVD scheme.
- The time discretization is performed by Strang-type fractional step method.
- Chemical reactions are treated by a point implicit way to avoid stiffness

→ **Petersen & Hanson model (High pressure model)**

High Pressure Hydrogen Leak from a Tube (4)

Numerical Conditions

- Reservoir tank pressures are 3.8, 6.8 MPa.
- Reservoir tank temperature is 300 K.
- The computational domain is rectangular.
- The left boundary condition is a solid wall and other boundaries are free stream conditions.
- A small hole of 10 mm diameter is located at the center of the left wall or tube inner diameter of 10 mm.
- The H2 jet is choked at the hole and the Mach number there is unity.
- The initial ambient air is at 0.1 MPa, 300 K, and at rest.
- The uniform grid size of dx=dy= 20 μm.

High Pressure Hydrogen Leak from a Tube (5)

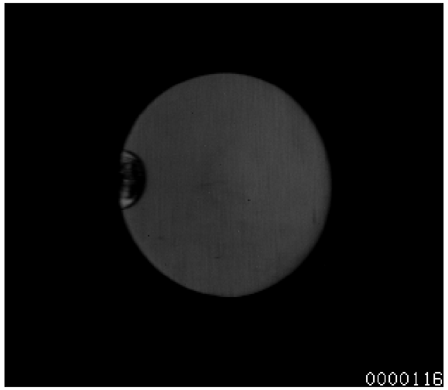
Numerical Conditions

- Reservoir tank pressures are 10, 40, 70 MPa.
- Reservoir tank temperature is 300 K.
- The computational domain is rectangular.
- The left boundary condition is a solid wall and other boundaries are free stream conditions.
- A small hole of 1, 3, 5 mm diameter is located at the center of the left wall.
- The H2 jet is choked at the hole and the Mach number there is unity.
- The initial ambient air is at 0.1 MPa, 300 K, and at rest.
- The uniform grid size of dx=dy=10 or 20 μm.

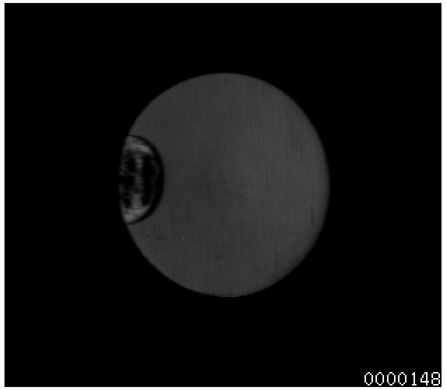
High Pressure Hydrogen Leak from a Tube (6)
-The case of the small hole-

High speed movie

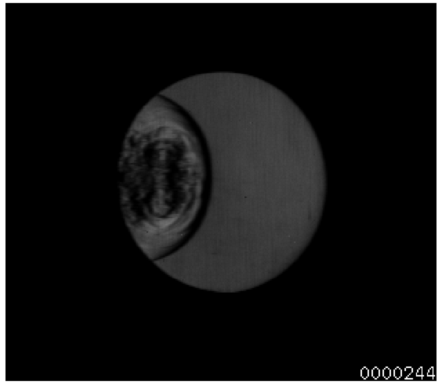
No ignition case



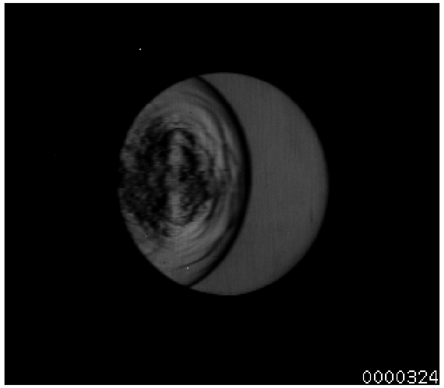
(a) 40 μ



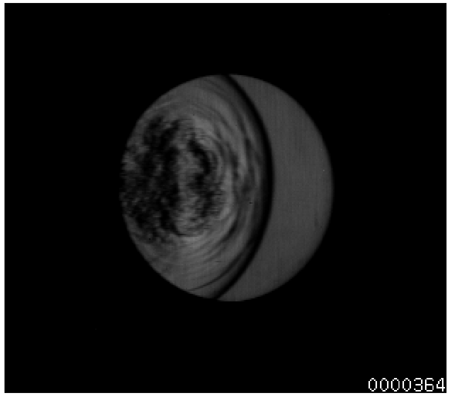
(b) 72 μ



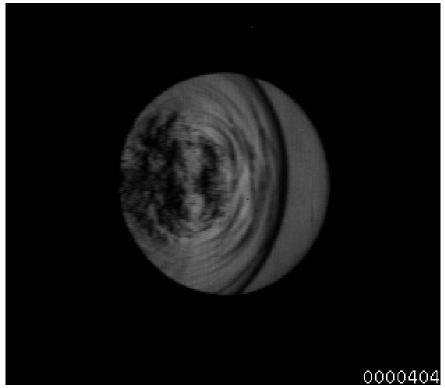
(c) 168 μ



(d) 248 μ



(e) 288 μ



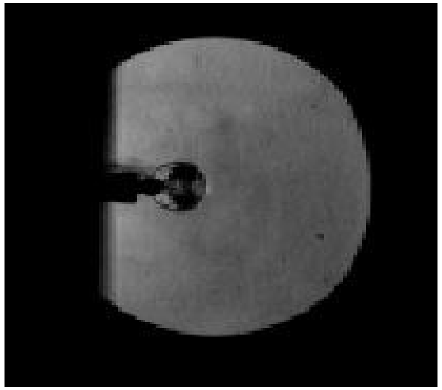
(f) 328 μ

- **The hole diameter is 10 mm. The hydrogen pressure is 2.3 MPa.**

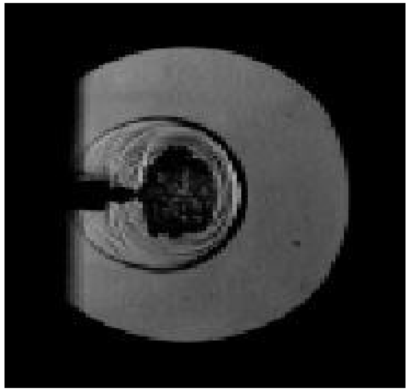
High Pressure Hydrogen Leak from a Tube (7)
-The case of the tube-

High speed movie

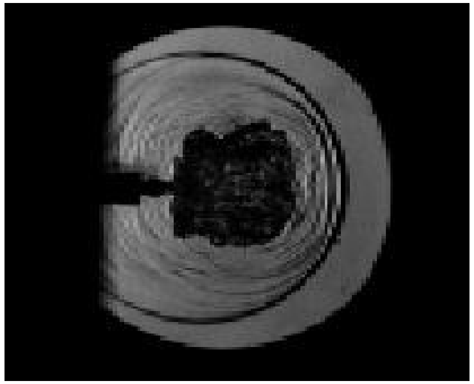
Ignition case



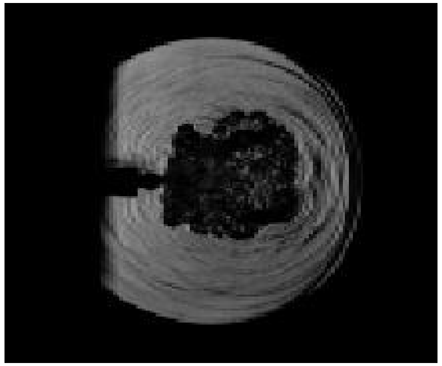
(a) 20 μs



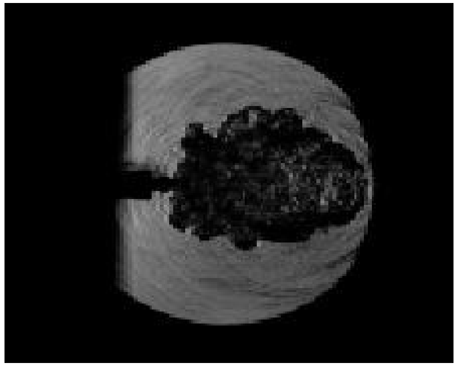
(b) 84 μs



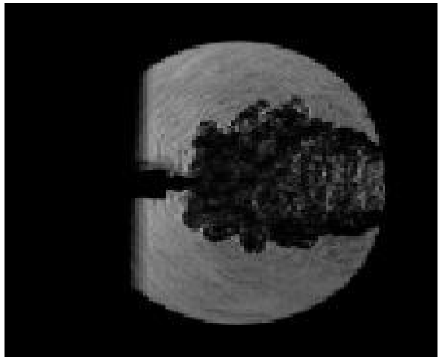
(c) 148 μs



(d) 196 μs



(e) 276 μs

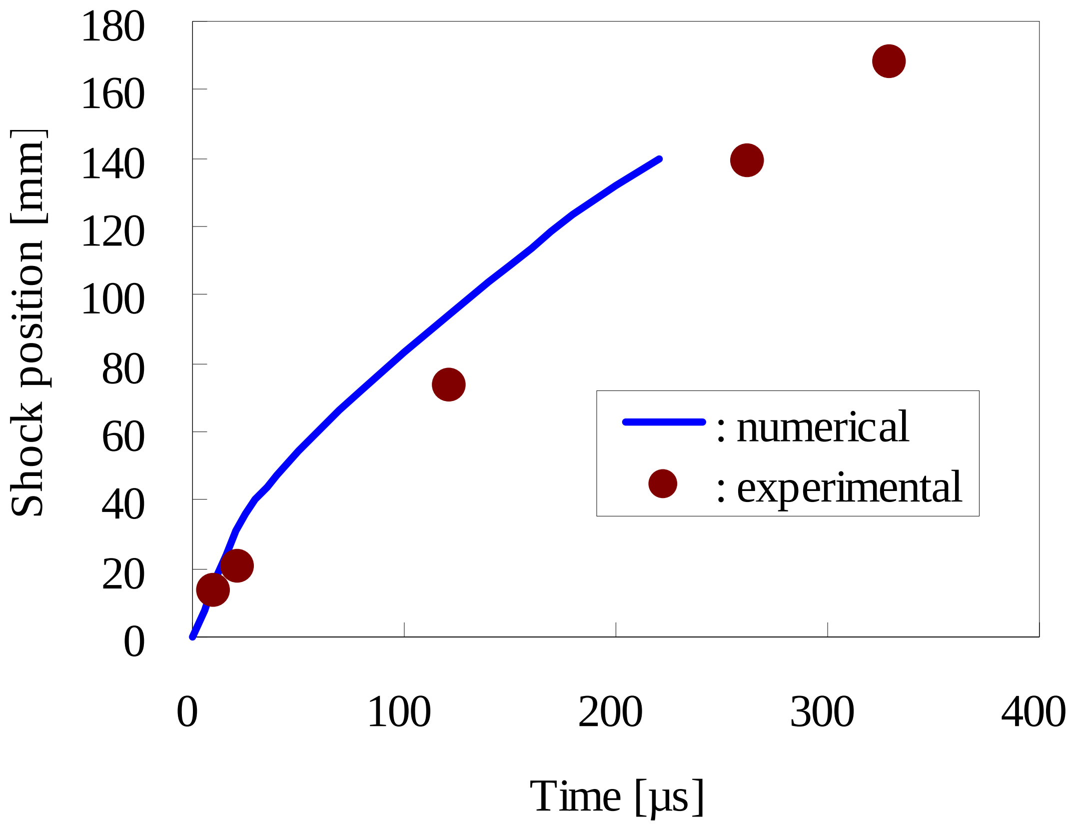


(f) 328 μs

- **The hole diameter is 4.8 mm. The tube length is 116 mm. The hydrogen pressure is 6.8 MPa.**

High Pressure Hydrogen Leak from a Tube (8)

-Comparison between experiments and numerical analysis-



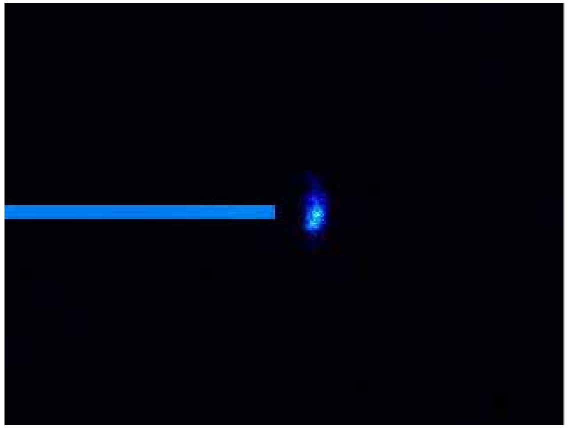
- H2 temperature is 671 K. H2 pressure is 3.8 MPa. Calculation does not include chemical reactions. The computation explains the experimental results well.

High Pressure Hydrogen Leak from a Tube (9)

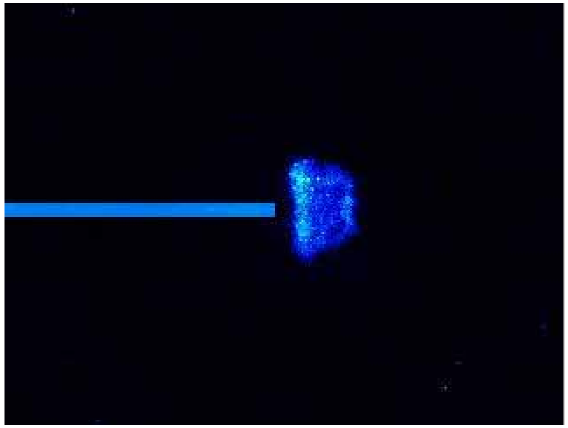
-OH emission pictures for the tube case-

Auto-ignition

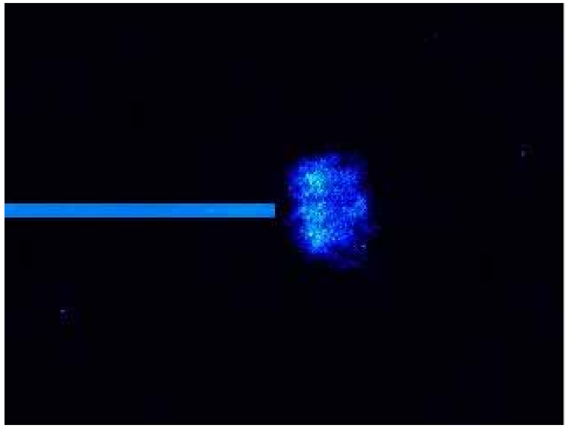
(a) 125 μs



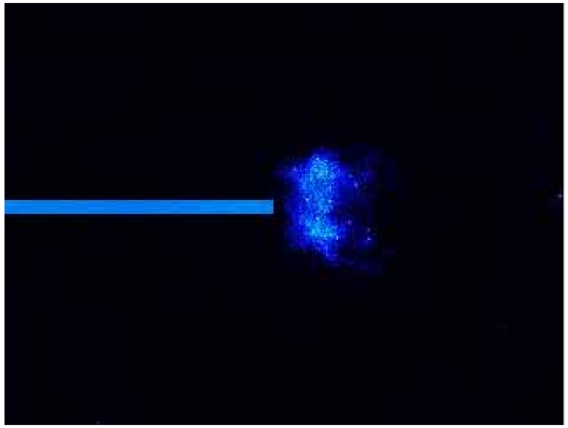
(b) 150 μs



(c) 175 μs



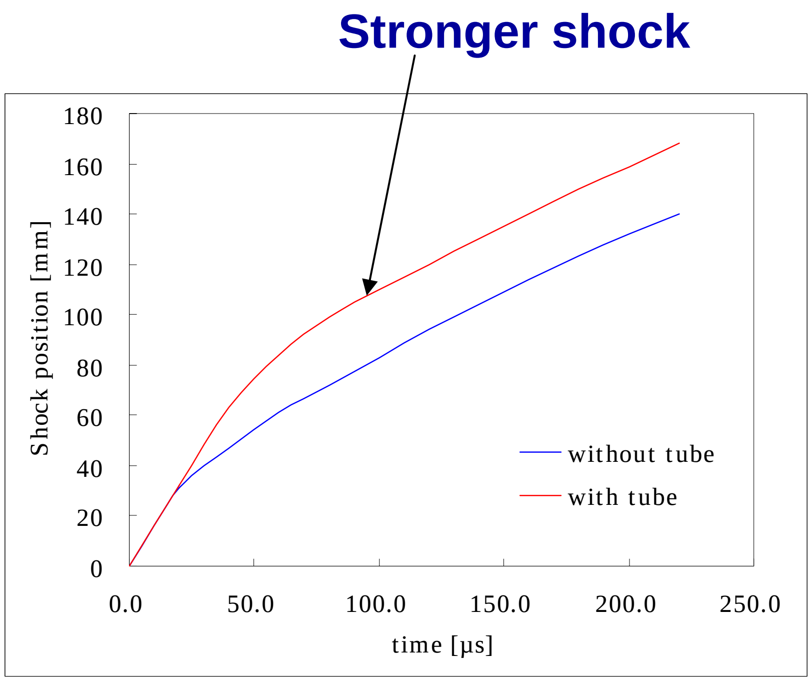
(d) 200 μs

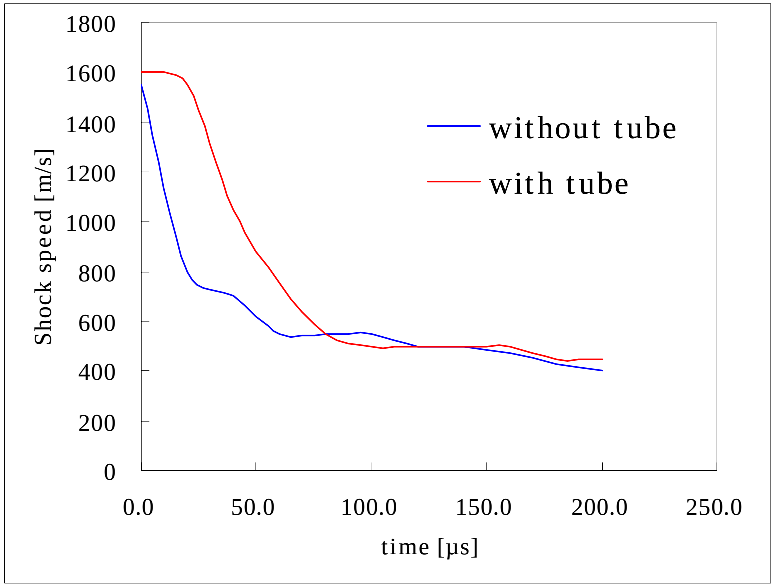


- **The tube diameter is 4.8 mm. The tube length is 115 mm. The flame (lifted flame) stays at the tube exit.**

High Pressure Hydrogen Leak from a Tube (10)

-Comparison between the jet from a hole and that from a tube-





(a) leading shock position

(b) leading shock velocity

- **The leading shock decays quickly for both a hole case and a tube case.**

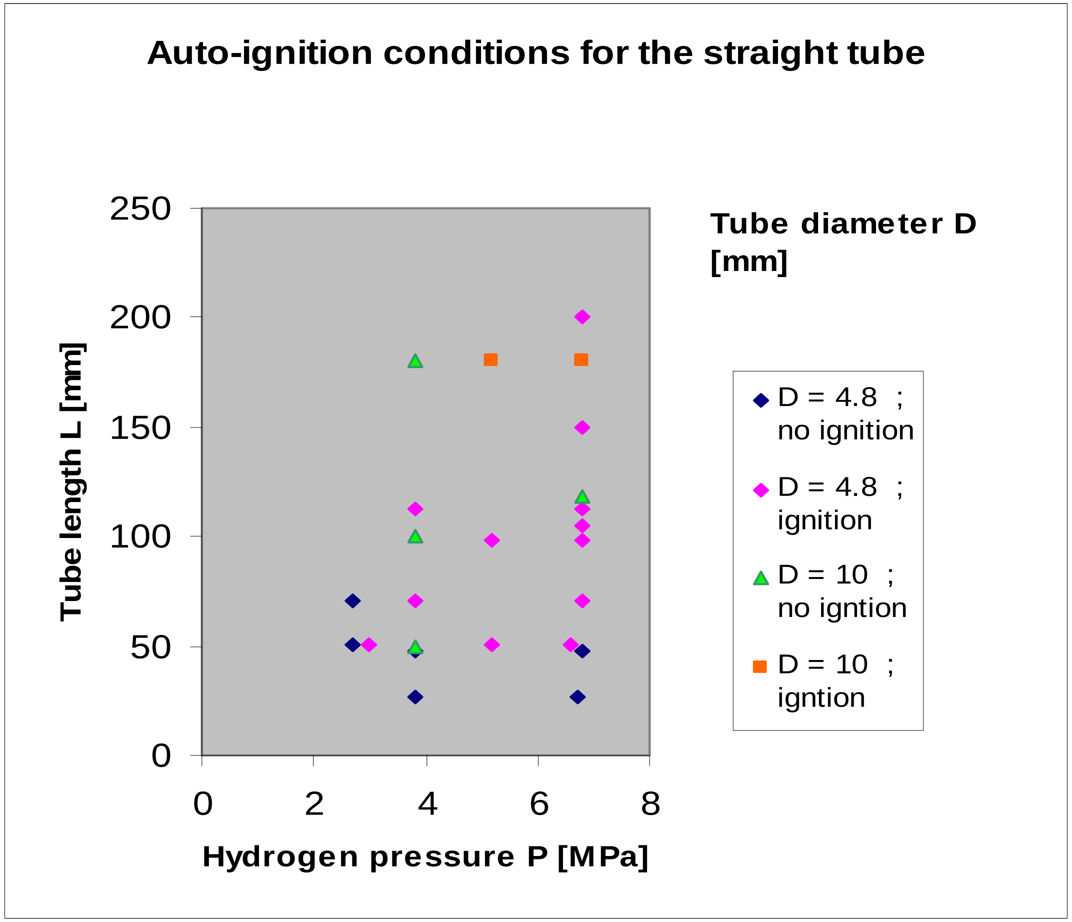
High Pressure Hydrogen Leak from a Tube (11)

-Auto-ignition conditions for tube cases-

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

- **The larger diameter tube and the higher hydrogen pressure conditions give hydrogen auto-ignition.**

High Pressure Hydrogen Leak from a Tube (12)
-Auto-ignition conditions for tube cases-



- **The longer tube length is also important factor for auto-ignition.**

High Pressure Hydrogen Leak from a Tube (13)

Summary

- Hydrogen jet coming out of a hole does not auto-ignite at the reservoir pressures up to 70 MPa for any size of hole dimeter.
- Hydrogen jet coming out of a tube has a different story: hydrogen auto-ignites depending on the tube diameter, tube length, and reservoir pressure.
- The hydrogen auto-ignites in the tube too.

Whole Summary

- Hydrogen leak from a hole may not auto-ignite for any reservoir pressure cases unless some special condition is set.

- However hydrogen leak will auto-ignite when it comes from a tube depending on tube diameter, tube length, and reservoir pressure.

- As far as we know, hydrogen auto-ignites inside of the tube if the tube has an oxidizer.