Hydrogen-air detonability limits in long tubes obtained using detonative driver gases

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Abstract

　The unstable phenomena near the lean detonability limits in atmospheric hydrogen−air mixtures are shown using 20−25−m−long test sections of 28, 53, and 105 mm inner−diameter gas−supply tubes. The driver section of the apparatus is charged with stoichiometric hydrogen−oxygen mixture to inject a detonation wave into the test section. Stronger equimolar acetylene−oxygen mixture and weaker 20 vol.% hydrogen−air mixture are also used as driver gases. Three propagation modes of deflagration, galloping detonation, and stable detonation are identified in quasi steady−state regimes. The detonability limits are 18 vol.% for the 28 mm ID tube, 15 vol.% for the 53 mm ID tube and 16 vol.% for the 105 mm ID tube. The results show that the pitch length of galloping detonations is about 10−15 m. This suggests that a longer test section is required to examine the self−sustainability of galloping detonation.

Keywords : hydrogen, detonability limit, self−sustainability

1. Introduction

　With the recent development of the utilization of hydrogen as a next−generation fuel, hazard evaluation of the explosive gas is an important industrial issue[1]. Since detonative explosion especially causes the most severe damages in comparison with other explosion phenomena, the critical conditions for detonation propagation in tubes are the most important concern. The lower detonability limit is usually defined as the critical fuel concentration below which self−sustained detonation wave cannot be established.

　There are two ways for determining the detonability limits in tubes. One way is observing the transition from deflagration to detonation. The transition through turbulent flame acceleration is usually enhanced installing obstacles, e.g. coiled wires. The other way is injecting a detonation wave of strong gas mixture in driver section into a test section of weaker gas mixture. In this experimental study, the latter method is used.

　In general, when strong driver gas is used, galloping detonation mode appears near the detonability limit between the stable CJ detonation and the flame propagation. The galloping mode shows an oscillatory behavior of the distance between the lead shock wave and the rear reaction zone. The shock wave and the reaction zone repeat a pair of amplification and decay phases. The status quo of our understanding does not provide an estimate of adequate observation tube length to determine whether the galloping detonation continues in a self−sustained mode or eventually decays. Observation of 2or3 cycles of the galloping oscillations is at least reasonable requirement for determining the detonability limits properly.

　Table 1 shows the lean detonability limits of hydrogen−air mixtures obtained using strong driver gas. These experiments are different in tube diameter, driver gas, and criteria for determining the detonability limit. Besides these differences, the more crucial concern is that the galloping detonation mode is not recognized. This strongly suggests that the tube length is not sufficient in these

Table 1   Lower detonability limits in the literature.

Diameter [mm]	Lower limit [H_2vol.%]	Test Section Length [m]	Refs.
14	18.3	9	2)
28	12	5.2	3)
54	11	5.2	3)
305	15	12.2	4)
430	13.6	12.2	5)

experiments.

More precise recent experiments using long tubes[6),7)] have revealed the detailed mechanism of the phenomena near detonability limits and the onset of detonations. The experimental study of low−pressure propane−oxygen stoichiometric mixtures near detonability limits[6)] using 24.5−m−long test section has shown that the local propagation velocity of galloping detonation varies 0.3—1.4 times of the CJ velocity around the mean value of 0.6 times of the CJ velocity with the pitch length of about 10m.

The aim of the present experimental study is to pursue the near−limit phenomena of atmospheric hydrogen−air mixtures using 20—25m test sections of a few different tube diameters. The test sections were connected to a driver section with a ball valve. The driver gas mainly used was stoichiometric hydrogen—oxygen mixture. Stoichiometric hydrogen—oxygen detonation transmits a shock wave with a velocity of 1500m s^{-1} into the test section, which corresponds to the typical hydrogen—air mixture detonation velocity.

2. Experimental Setup

We used an experimental apparatus similar to that used in the experiments by Matsui[3)]. Figure 1 is a schematic diagram of the present detonation tube.

The effect of tube inner−diameters of 28, 53, and 105 mm on the detonability limit is examined in the test section of 20—25m long, in which self−sustainability of detonations is carefully examined. The test sections consist of several seamless tubes and a 1—2m tube with M18 holes for installing ion probes and pressure transducers. The ratio of the length L to diameter D, L/D, ranges from 190 to 850 under different conditions of the experiments. An oscilloscope (Yokogawa DL750) records the outputs of pressure transducers and ion probes. The oscilloscope can simultaneously record 16 analog inputs

and 16 logical inputs. An analog input is digital data with 16 bits for 1 M samples per second or 12 bits for 10M samples per second. For pressure records, analog signals from PCB 113A24 pressure transducers are used. Near the ball valve in the driver section, the pressure signals are recorded to ensure that fully developed CJ detonations are established. The length of the driver section is 1 or 2m. At the end of the driver sections of 53 and 105 mm diameter tubes, a 28−mm−diameter tube is added to easily establish driver detonation wave. Ion probes are charged at 1—3V and each connected to a detection resistance of 100kΩ. Analog operational amplifiers (TA75358P) amplify the ion probe signals 1000 times, and the amplified signals are stored in logical record. All gas mixtures were prepared at least 6 hours prior to the experimental run in bottles employing the partial pressure method.

The experimental procedure is as follows.

1. Open the ball valve connecting the driver and test sections. Then draw air, residue gases and water vapor until the pressure is attained below 1 kPa in the whole tube.
2. Fill the test gas of the hydrogen−air mixture to the whole tube.
3. Close the ball valve and draw the test gas mixture from the driver section again.
4. Charge the stoichiometric hydrogen−oxygen mixture to the driver section.
5. Immediately after opening the ball valve, ignite the driver gas mixture with an electric spark from a 5000 pF capacitor charged at8kV.

3. Results and Discussion

Table 2 summarizes the experimental results. Haloua et al.[6)] classified propagation behaviors into four categories ; i.e., stable, stuttering (spin), galloping detonations and a fast flame, based on the continuous detailed records of microwave Doppler velocimetry of local wave velocity. Since stable and stuttering detonation modes have almost CJ velocity on average, they are not distinguishable in this experiment. Certain quantitative criteria are set for classifying the observed phenomena. 'Attenuate' in the table corresponds to the cases in which the shock wave overpressure is less than 1/10 of the CJ overpressure and the average propagation velocity is less than 600m s^{-1} in the rear 5m length of the test section. This is a failure of detonation initiation and corresponds to fast flame. 'Gallop'
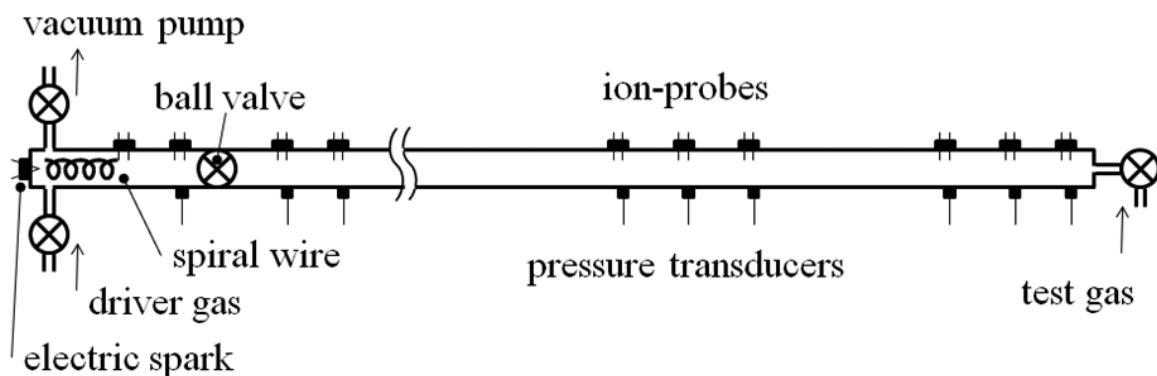

Figure 1   Schematic diagram of the detonation tube.

Table 2   Experimental results of detonation initiation by another detonation as a driver. Each mark shows one experimental result. The stoichiometric mixture of hydrogen and oxygen was used as driver gas except the thick mark with an equimolar mixture of acetylene–oxygen and the thin mark with a 20 vol.% hydrogen—air mixture.

Hydrogen concentration [vol.%]		10	11	12	13	14	15	16	17	18
Tube diameter [mm]	105	✕	×	⊗△	△△	△△	△△	○○	△△	○
	53		⊗	△△	×⊗×	△△	○△	○○	○	○
	28			⊗⊗⊗	△△△△		△△	△△	△△	○○

○ Stable △ Gallop ✕ Attenuate   Thick: strong driver Thin: weak driver

corresponds to the cases in which transition occurs in the middle part of the test section or a shock wave remarkably detached from the flame. In these cases, the average velocity is less than the CJ velocity by tens of percentage. However, there is an overdriven state at the transition where the propagation velocity exceeds the CJ velocity over a short range. The oscillatory time–behaviors of wave velocity is not obtained in some experimental runs, since the galloping pitch length is larger than 10–15m. In the present study, the lower detonability limit is determined as the lowest fuel concentration above which stable detonations appear. We classify the galloping detonation into subcritical initiation phenomena in the present study, since there remains a possibility that the galloping wave decays gradually to deflagration in several oscillatory propagation cycles.

3.1 28 mm tube

The stable detonation limit was 18 vol.% and the galloping detonation limit was 13 vol.% in the present 28 mm tube experiments. These limits are clearly determined irrespective of the driver gases without any scattering, e.g., mixed go and no-go results at the same fuel concentration. Figure 2 includes the pressure records of 12 vol.% hydrogen—air mixture into which an equimolar acetylene–oxygen detonation is injected and show an overdriven detonation in the early stage. The figure also shows the temporal positions of a flame (reaction front) and an attenuating shock wave.

Channels 2 to 5 are at intervals of 5m in the test section. The results show typical attenuation, but the propagation velocity exceeds 1000m s^{-1} in the early stage. Since we used a relatively long tube, we could observe the pressure
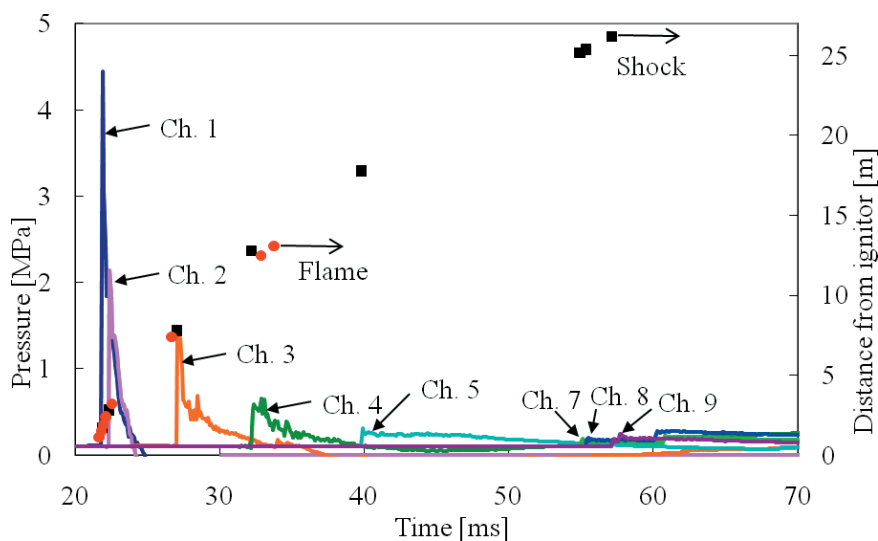

Figure 2   Pressure records and time–position diagram of the experiment in a 28 mm tube with a 12 vol.% hydrogen–air mixture in the test section and an equimolar acetylene–oxygen mixture in the driver section. The ball valve is located between the pressure transducer Ch. 1 and Ch. 2.
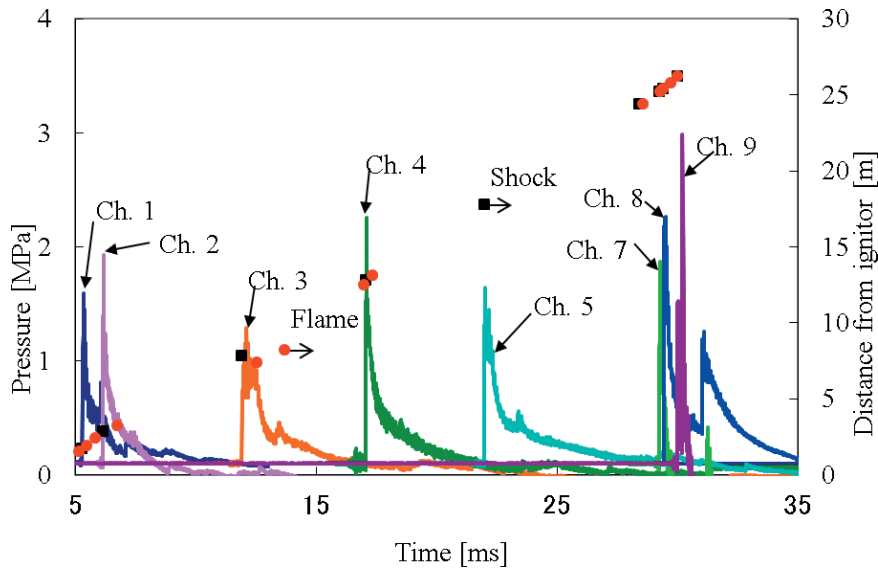
Figure 3   Pressure records and time–position diagram of the experiment in a 28 mm tube with a 13 vol.% hydrogen–air mixture in the test section and a 20 vol.% hydrogen–air mixture in the driver section. The ball valve positioned between the pressure transducer Ch.1 and Ch.2.
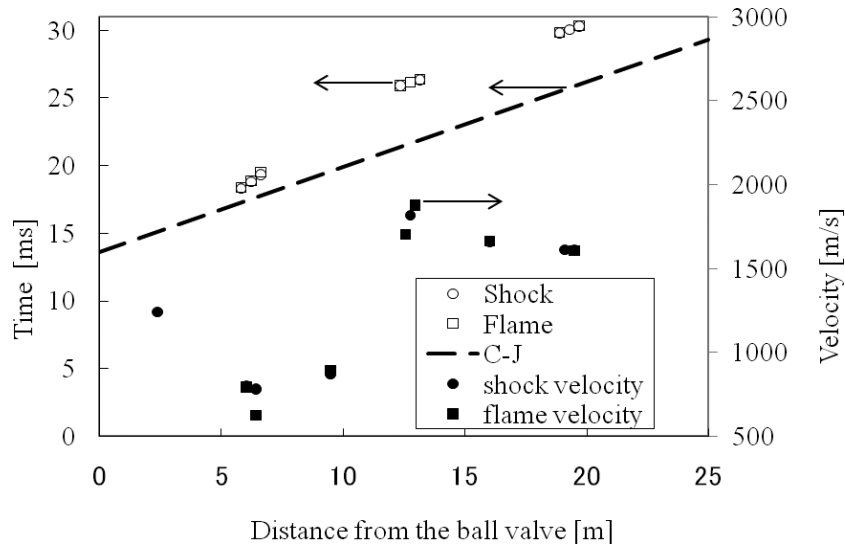

Figure 4 $x–t$ and $x–V$ diagrams for 17 vol.% hydrogen–air mixture in the 105 mm tube. The calculated velocity of CJ detonation with 17 vol.% hydrogen–air mixture is 1594 m/s.

decay of the 12 vol.% mixture. This decaying stage is not observed in Matsui's experiments using shorter tubes (see Table 1). The pressure records of channels 3 and 4 show an insufficient catch–up of the reaction front (second pressure peak) to the lead shock wave which results in a failure of detonation reinitiation. The average shock velocity between channels 8 and 9 is 383m s^{-1}. The shock wave and the flame separately propagate at their characteristic velocities to provide a typical deflagration pressure distribution.

Figure 3 shows the records of a typical galloping mode in hydrogen 13vol.% mixture. Around Ch. 3, an insufficient catch–up of the reaction front to the shock also can be seen. However, they reunite around Ch. 4. Two cycles of the oscillatory behaviors are observed, and the galloping pitch length is about 10–15m.

3.2 53 mm tube

In the 53 mm tube experiments, the results scatter in hydrogen 13vol.% mixture experiments. We could not find the reasons for the sequential change of gallop–flame–gallop in 12, 13, and 14 vol.% mixtures. We determine the detonation limit as 15 vol.% and the galloping detonation limit as 12 vol.%. The dependence of the limit on the tube diameter strongly suggests the wall effects. There are two effects of the tube wall. The first is the boundary layer effect, which is suppressive to detonation propagation. This effect increases for smaller tube diameter. The second is the reflection of transverse waves in the lead shock surface, which strengthens the shock triple points to enhance rapid heat release. This second effect enhances the detonation propagation and increases for smaller tube diameter. These two effects are opposite and compete with each other, and the net of the two effects varies with
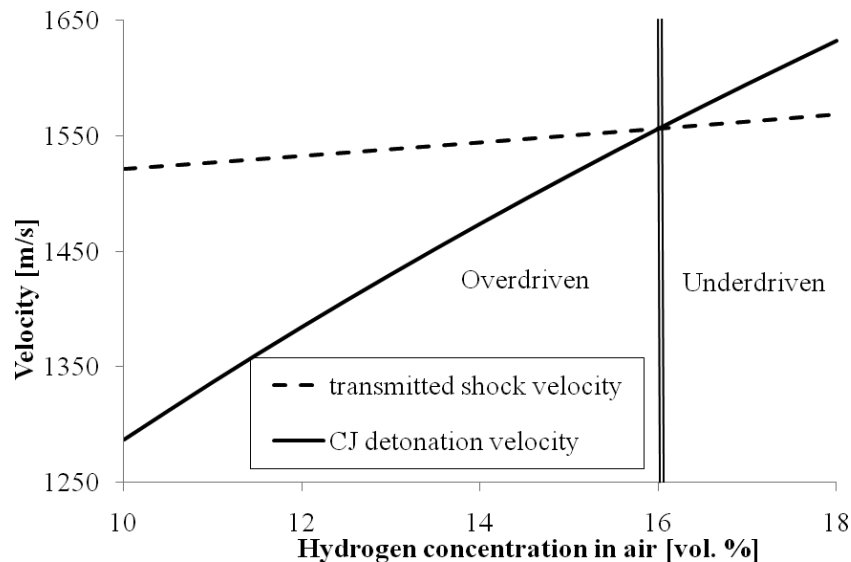
Figure 5 Velocity of CJ detonation and the transmitted shock wave from hydrogen−oxygen detonation to hydrogen−air in SATP at each concentration.

tube diameter. Therefore, the optimal tube diameter exists for providing the lowest fuel concentration of detonability limit. In the present experiments, 53 mm tube provides the lowest limits of both the gallop and stable modes in comparison with the results of 28 mm and 105 mm tube experiments. The detonability limit of 11 vol.% obtained in Matsui's experiments in Table 1 is close to the galloping limit 12 vol.% in the present experiments. Together with the results of 28 mm tube experiments, the present experiments conclusively show that short test sections cannot provide the galloping mode.

3.3 105 mm tube

The stable detonation limit is 16 vol.% and the galloping detonation limit is 12 vol.% in the 105 mm tube experiments. The results of 17 vol.% hydrogen mixture in Figure 4 show an exceptional initiation mode in which the lead shock wave and the reaction front closely interact with each other and propagate as a detonation wave. The local velocity of the wave at the location about 6−7 m slows down to 800 m s^{-1}, and the estimated shock−flame induction distance is about 15 cm corresponding to the induction time of 0.183 ms at the maximum. The wave accelerates in the distance of 7−13 m of the test section to propagate at CJ velocity in the distance of 13−20 m.

To see the detail of the phenomena of 16 and 17 vol.%, we calculated transmitted shock wave velocities as shown in Figure 5. The CJ detonation properties can be calculated using CEA code[8]. The cell size of the stoichiometric hydrogen−oxygen detonation is sufficiently small to consider that the detonation wave is planar in the driver section. Therefore, the shock wave relation can be applied to the contact surface between the driver and test gases. The transmitted shock wave in mixtures with concentrations of 16 vol.% and lower is in an overdriven state relative to the CJ condition. Therefore in 16 vol.%, transition is not necessary. In contrast, in the case of mixtures with concentrations of 17 vol.% and higher, the shock wave is in an underdriven state. The transition

occurred from the underdriven state in 17 vol.%, but it took relatively long distance and time. Then the velocity in the rear 5 m length of the test section is almost the same as the CJ velocity of 17 vol.%. The longer test section is necessary to confirm that the another oscillation will appear or not in 17 vol.%.

4. Summary

Through the experiments using long tubes and stoichiometric hydrogen−oxygen driver gas, we conclude that the minimum concentrations required for initiating self−sustained detonation waves are 18 vol.% for the 28 mm tube, 15 vol.% for the 53 mm tube and 16 vol.% for the 105 mm tube. To judge the self−sustainability near the limit conditions with 105 mm tube, more experiments with different compositions of the driver gases and longer test sections are necessary.

Acknowledgments

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爆ごうドライバーガスを用いて得た長い管内の
水素－空気爆ごう限界

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　大気圧下での水素－空気混合気の爆ごうの下限界について，直径28，53，105mm，長さ20～25mのテスト部分を持つ管における実験を行った．定常爆ごうをテスト部分に送りこむため，ドライバ部分に量論比の水素－酸素混合気を導入して用いた．また，より強いドライバとして等量のアセチレン－酸素混合気や，より弱いドライバとして20 vol. ％の水素－空気混合気も用いた．爆燃，ギャロッピング爆ごう，定常爆ごうが観測された．爆ごう限界は，28mm管で18vol.%，53mm管で15vol.%，105mm管で16vol. %である．結果からギャロッピング爆ごうでは10～15mの間隔で再転移が起こるため，より長いテスト部分が必要であることがわかった．

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