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Experimental study of detonation limits in methane-oxygen mixtures:

Determining tube scale and initial pressure effects

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Abstract

In this paper, detonation limits in stoichiometric methane-oxygen mixtures with varying tube inner diameter and initial mixture pressure were investigated. Detonations in tubes with different inner diameter (D = 36 mm, 25 mm, 20 mm and 13 mm) and low initial pressure from 3.5 ~ 18 kPa were studied. Smoked foils were applied to observe the evolution of the detonation cellular structure for various initial conditions. An alternate length scale at the limits is examined, L_{dcs}, which is the maximum length from the beginning of the test section after which cellular patterns can no longer be observed. Simultaneous local velocity measurements were obtained by photodiodes to complement the L_{dcs} results. The study also aims to reveal relation between the near-limit detonation dynamics, the tube geometry, and the thermodynamic properties of the mixture. Past the failure limit, L_{dcs} decreases with decreasing initial mixture pressure for a given tube diameter, and L_{dcs} decreases faster in a smaller diameter tube. In the D = 13 mm tube, galloping detonation mode is observed, and the length of the galloping cycle is reduced with an increase in initial pressure. To further characterize the onset of detonation limits, a scaling analysis of L_{dcs} with tube inner diameter (D) and detonation cell size (λ) was performed. The experimental results show that the decrease of L_{dcs}/D and L_{dcs}/λ are more abrupt in smaller diameter tubes with decreasing initial pressure. At low initial pressure, the boundary layer displacement thickness growth is significant in the flow structure. Since the distribution of global curvature over the whole detonation front is faster in smaller tube, it thus leads to a more abrupt decrease sensitive to initial pressure. For increasing pressure closer to the critical failure limit, the boundary layer displacement thickness is becoming less comparable to the tube diameter. The failure mechanism appears to be more dominant by the rate of transverse waves attenuation or cell disappearance. Lastly, by comparing the detonation cell size and the tube scale at the critical limits condition in different tubes, $\lambda = \pi \cdot D$ is shown to be an appropriate limit criterion of detonation propagation in agreement previous studies.

Keywords: Detonation limits; Tube inner diameter; Initial pressure; Detonation cellular structure

1. Introduction

An explosion is the result of a rapid expansion of product gases, for example, from a combustion process. Explosion is a high energy release process and it creates a blast wave, whether this wave decays to sub- or supersonic depends on the amount of energy released. In the subsonic case, explosions are created by the ignition and subsequent slow burning of combustibles known as deflagration. In the supersonic case, explosions result when the combustion occurs in the detonation regime. A detonation is a supersonic, combustion-driven, compression wave. Ideally, the detonation structure is described by a lead shock followed by a reaction zone consisted of an induction followed by the exothermic energy release. It is a supersonic combustion with an overpressure of about 20~30 times the initial pressure and a propagation velocity of about 2~3 km/s. Real detonations have a three dimensional cellular structure, this complicated structure comprises of transverse waves, triple-points, and turbulent shear layers [1-3].

For the past century, investigations on detonation dynamics have attracted numerous researchers mainly due to two aspects. First is to prevent and mitigate the formation of detonations in industrial processes. From a safety point-of-view, only by knowing the critical conditions that the detonation initiation or failure then can the corresponding safety precautions be effectively formulated [4]. The second aspect is to develop potential detonation-based propulsion systems and harnesses their high propulsive efficiency. Examples include pulse detonation engines [5-8], rotating detonation engines [9-11] and oblique detonation wave engines [12-19].

Soon after the discovery of the detonation phenomenon, Chapman and Jouguet proposed a theory that quantitatively calculated the detonation velocity of a combustible mixture, known as

Chapman–Jouguet (CJ) theory. However, the CJ theory suggests that the detonation velocity depends only on the thermodynamic properties of the mixture, but independent of detonation wave structure and boundary conditions. Therefore, for a detonation propagating in a given mixture and confining environment, the self-sustained propagation state can always be maintained regardless of the tube geometry and other boundary conditions. In reality, however, the propagation of detonations is strongly affected by the boundary conditions; this is owing to the detonation vulnerable to boundary effects because of its finite thickness of the reaction zone [20]. In general, the boundary effects result in a velocity deficit and further render the detonation to decay, and finally approach the detonation limits leading to failure.

Table. 1 Previous literature of detonation propagation limits

No.	Authors	Year	Tube length/m	D or H /mm	$(L/D, L/H)_{max}$
1.	Ishii & Grönig [21]	1998	14	45	311.1
2.	Haloua et al. [22]	2000	24.5	38	644.7
3.	Ishii et al. [23]	2002	5.05	50.5	100
4.	Chao et al. [24]	2009	3	CT:65, AC:2.2/6.9	1363.6
5.	Kitano et al. [25]	2009	3	3/6/10	1000
6.	Camargo et al. [26]	2010	3	1.8/ 6.3/ 9.5	1666.7
7.	Ishii & Monwar [27]	2011	5.05	50.5	100
8.	Sadahira et al. [28]	2013	5	3.1/ 6.0/ 8.8	1612.9
9.	Lee et al. [29]	2013	4.16	CT:13/44/65, AC:3.175/6.35/9.525	1310.2
10.	Gao et al. [30-34]	2014-2016	2.438/4.118	1.5/3.2/12.7/31.7/ 50.8	2745.3
11.	Wu & Lee [35]	2015	3	50.8	59.1
12.	Yoshida et al. [36]	2016	6	5.8/8/10	1034.5
13.	Jackson et al. [37]	2016	30	4.1	7317.1
14.	Wang et al. [38]	2017	2.5	32	78.1
15.	Zhang et al. [34, 39-50]	2015-2019	2.5	CT:4/14/36, AC:2/4.5/7	1250

Detonation limits is a fundamental dynamic parameter of detonations [39, 40, 43, 48, 51]. As the detonation limit is reached, a variety of unstable propagation phenomena occur, e.g.,

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galloping detonations, spinning detonations, and stuttering detonations. Detonation limit conditions are reached through reducing the initial pressure of the combustible mixture, changing the confinement geometry and/or scale, increasing the amount of an inert diluent, changing the roughness of the confinement boundary, etc. For reference, a number of investigations regarding detonation limits in recent years are tabulated in Table.1.

Although similar detonation tubes with circular or annular geometries have been normally used, the tube configuration (e.g., tube length, or the ratio between the length and inner diameter L/D) is quantitatively different (as shown in Table.1). Therefore, the critical condition (e.g., p_c - critical pressure) of detonation propagation limit for each apparatus also varies. For example, Wu and Lee [35] reported the maximum initial pressure of $CH_4 + 2O_2$ mixture at which spinning detonations failed was 4.3 kPa, in which they employed a polycarbonate tube of 50.8 mm in diameter and 3 m long. However, a pressure value of 3 kPa was found by Zhang et al. [46] for the same mixture, but in a tube of 36 mm in diameter and 2.5 m long. Generally, the detonation limit is preceded by a detonation velocity deficit. Previous studies [34, 47, 52] have suggested that the mechanism of the velocity deficit can be attributed to the flow divergence that caused by the boundary layer effect in small tubes. Besides, the velocity deficit can also be caused by the effect of momentum and heat losses to the wall [24, 25]. Therefore, it is reasonable to speculate that the critical pressure for self-sustained detonations in tubes with different geometries should also be different. To this end, a universal criterion of detonation limits that considers the tube geometry and initial mixture conditions is desirable.

For a given duct, as the initial pressure is well above the detonation limit, the wave can self-sustainably propagate, and its velocity is usually steady. The detonation limit can be reached though gradually reducing the initial pressure, whereby the cellular structure vanishes

completely after a distance from the beginning of the tube, and the detonation velocity starts to

decay. With deceasing initial pressure, the combustion mode turns into a fast deflagration

regime. On the other hand, for a given mixture and initial pressure, detonations are more

affected by smaller diameter tubes, and the detonation wave fails after a shorter distance if the

tube diameter is reduced. Although the above behaviors can be qualitatively presumed,

nevertheless, the quantitative relationship between the detonation propagation distance with the

independent effects from tube inner diameter and the initial pressure of mixture are interesting

to be investigated. In industry that involves the use of chemical processes, this quantitative

relationship of detonation limit with those factors is important for the designers to take effective

measures to prevent the formation or transmission of a detonation in pipes and ducts. In addition,

this quantitative relationship is also meaningful for the engineers that investigate the

detonation-based propulsion devices, in which the detonation is expected to be maintained and

has to be away from the limit conditions to keep the thrust ability as an engine.

In this study, detonation experiments were performed in tubes with four inner diameters

(i.e., D = 36, 25, 20 and 13 mm). The detonation cellular patterns and wave velocities were

simultaneously recorded at varying initial pressures. In order to examine the results, an alternate

length scale is considered. The maximum length of detonation cellular structure (L_{dcs}), defined

as the length from the start of the test tube section to the location where no detonation cellular

pattern is observed, was obtained for each condition to explore the quantitative effects of tube

geometry and the thermodynamic properties of the mixture on the detonation limit. It is worth

noting that similar characteristic length was also used in Radulescu and Lee [53] to define the

failure limit in porous walled tubes. They reported a failure length, and this length can be

approximately considered a constant, of the order of 3 to 7 λ. The failure is mainly due to the

145 losses and mass divergence into the porous walls and not inherently by the tube scale or

146 geometry. The porous walled tubes not only attenuate transverse waves and the global mass

147 divergence into the permeable walls introduces curvature slowly disturbed in the leading front.

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149 **2. Experimental Details**

150 **2.1 Experimental apparatus**
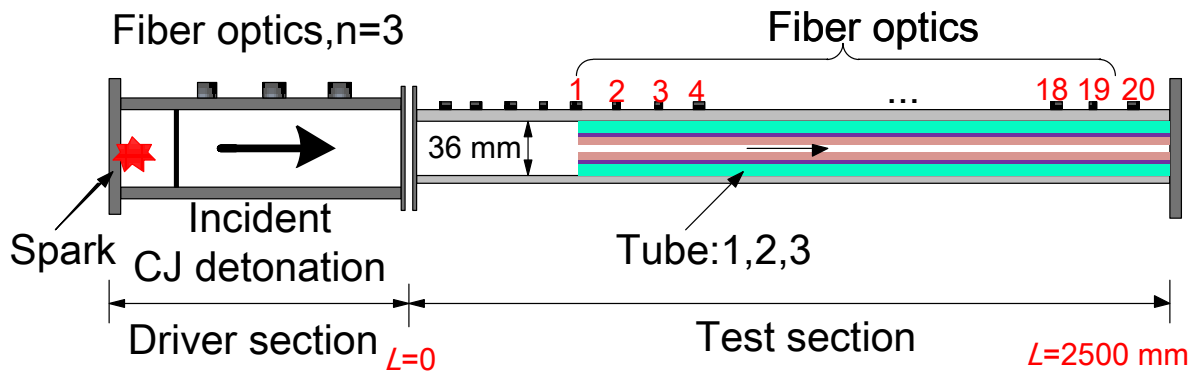
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152 Fig.1 Schematic diagram of the experimental device

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154 Detonation limits experiments were performed in a detonation tube facility 3700 mm in length.

155 It comprised both driver and driven sections, a diaphragm ($L = 0$) was used to separate these two

156 sections. The driver section is 1200 mm in length and 68 mm in inner diameter, the test section is

157 2500 mm in length and 36 mm in inner diameter, as shown schematically in Figure. 1. Equi-molar

158 $C_2H_2 + O_2$ was used as driver mixture, it is very sensitive and readily forms a CJ detonation, and the

159 initial pressure was 10 kPa. The test section was filled with the desired test mixture ($CH_4 + 2O_2$).

160 Various transparent acrylic glass tubes with smaller diameter were inserted into the test section,

161 which were used to change the test section's inner diameter. The inner diameters (D) of the tubes

162 (No. 1, 2, 3) were 25 mm, 20 mm and 13 mm, respectively. All the glass tubes were 2000 mm in

163 length (i.e., from $L = 500$ mm to $L = 2500$ mm). The initial pressure and tube diameter were two

important parameters that affect the detonation propagation limits. The initial pressure affects the characteristic scale of the detonation structure, while the tube diameter is related to the physical length scale of the boundary. The initial pressure is also a controlling variable to approach the detonation limits. In this study, a certain discrimination of tube inner diameter was required, i.e., from large (36 mm) to small (13 mm). The sizes are chosen such that the critical pressure for the limits will be within the range of safety for the present detonation facility. The selection of initial pressure was mainly dependent on the width of detonation cellular structure. In some cases, as the p_0 is relatively high, an entire soot foil recorded with cellular structure can be observed, indicating the condition is well within the detonation limit. Hence, any higher initial pressure is irrelevant for the present investigation on detonation limits.

In this study, L_{dcs} refers to the length from the start of the test section to the location where no detonation cellular pattern is observed. As shown in Fig. 1 the test section comprises two parts, the first part is a short length section ($L_1 = 500$ mm) prior the glass tube. The purpose of this section is ensure a well-established detonation is formed in the test mixture. Without this part, the detonation in the entrance of small tube could be from the driver mixture. To eliminate this possibility, this buffer section is necessary. The second part is the small glass tube section ($L_2 = 2000$ mm). In fact, it is possible in the definition of L_{dcs} to eliminate the constant length $L_1 = 500$ mm since the detonation only fails in the small glass tubes for all test conditions. Thus, all the L_{dcs} values will be simply shifted by a constant and such consequence will not affect the conclusion of this work. However, there are two reasons why L_1 is kept in the L_{dcs}. First, both L_1 and L_2 sections were filled with the same mixture at which a detonation propagates and that the detonation in the small tube is not directly initiated, but rather it transmits from a bigger inner diameter tube. In other words, the detonation has already established before the entrance of small tube, hence the detonation

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propagation distance before it goes into the small tube should be considered. Second, for completeness, the inclusion of L_1 in the present L_{dcs} definition can take into account possible losses in this first section particularly for cases with very low initial pressure conditions. Nevertheless, the latter may not be as significant since limits are reached far before in the small glass tubes.

It is also worth mentioning that, to minimize any shock wave reflection of the detonation front from the step change as it enters into a reduced small area, Xiao et al. [54] used a protruded rounded tip at the entrance. In this study, the area step change is not as severe as in [54]. It is also challenging to modify smoothly the entrance of the small tubes section. Hence, the transparent acrylic glass tubes used in this study are with a flat straight cutting edge. Although this configuration unavoidably results in shock reflection at the entrance of acrylic glass tubes and that the shock reflection may affect the early initial transient of detonation behavior in the small tubes, it provides a consistent initial condition for all tests. To significantly avoid any shock reflection resulted from the step change, a long gradual rounded ramp must be used. However, the latter will generate even much different initial conditions for each test as it allows the cellular detonation to adjust to different structure. Any roughness from any imperfect modification will also perturb the detonation entering the small tubes.

In the experiment, cellular detonation pattern was recorded by smoked foil technique. To obtain records of the cellular pattern in the test section, the foil was placed along the internal face of tube (typically L = 500 mm - 2500 mm).

In addition, the combustion wave velocity was also determined to observe the attenuation of the detonation propagation. The local wave velocity was obtained by computing the length over two adjacent arrival time signals; these signals were generated from optical fibers together with photodiode. In the test section, twenty-four fibers equally distributed, and the distance between each

other was 10 cm. Figure 1 shows 20 optical fibers from $L = 500$ mm to 2500 mm, and 4 more fibers were placed before $L = 500$ mm.

2.2 Experimental uncertainty

The uncertainties of experimental measurement were from two aspects: a) initial pressure measurement, and b) the determination of the length (L_{dcs}). Methane and oxygen at equivalence ratio $\varphi=1$ was used in this work, and $T_0=300$ K. The mixture was prepared by the partial pressure method in a 40-L high pressure mixing bottle and allowed to mix by diffusion for at least 24 h to ensure the homogeneity prior to be used. The bottle was initially evacuated to an absolute pressure of 0.1 kPa. It was then filled with the fuel (i.e., methane) to the desired pressure. For the stoichiometric condition, i.e., $\varphi = 1$ ($CH_4:O_2 = 1:2$), the mixing bottle was filled with the fuel to 60 kPa and afterwards, with oxygen into the bottle up to the 180 kPa desired initial mixture pressure in the mixing bottle. The gas handling was monitored by the OMEGA PX309 pressure gauge. Before each experiment, the detonation tube was first evacuated and then filled with the mixture from the high pressure mixing bottle directly. The initial pressure in the mixing bottle and detonation tube was measured by OMEGA pressure gauges (shown in Table 2). Table 2 gives the accuracy and maximum error of each gauge.

Table.2 The accuracy and maximum error of sensors

Type	Range/PSI	Accuracy	Maximum Error/ kPa
PX309	0-200	± 0.25%	± 3.45
HHP242	0-30	± 0.10%	± 0.20

Besides the pressure monitoring, uncertainties in smoked foils measurement also arose due to the unstable nature of detonation waves. It has been established that detonation waves are

essentially unstable and possess a transient 3-D structure, even though the global velocity is still

close to the one-dimensional steady CJ velocity. Based on the regularity of the detonation cellular

pattern and the stability parameter χ, Radulescu [55] and Ng [56] classified qualitatively various

mixtures into several types, i.e., stable, mildly unstable and highly unstable. Accordingly, $CH_4 +$

$2O_2$ is one of the typical unstable mixtures and has very irregular cellular pattern. Note that χ is a

dimensionless parameter, which is used to characterize the detonation stability. For unstable

mixtures, their values of χ are found to be much larger than stable mixtures (e.g., highly argon

diluted mixtures). For the latter mixtures, their cellular pattern are regular.

The measurements of L_{dcs} were conducted in different tubes. Five shots were performed at the

same initial condition for the $CH_4 + 2O_2$ mixture, i.e., $D = 25$ mm, $p_0 = 10$ kPa. The smoked foils (1

m in length) with the same thickness (0.1 mm) were inserted from $L = 1500$ mm to 2500 mm (See

Fig. 2). From Fig. 2, the longest distance L_{dcs} was 2500 mm, and the shortest distance was 2000 mm,

with an average value of 2220 mm, standard error of 86.02 mm ($\pm$ 3.87%). The statistics of

experimental uncertainty was illustrated in Fig. 3.
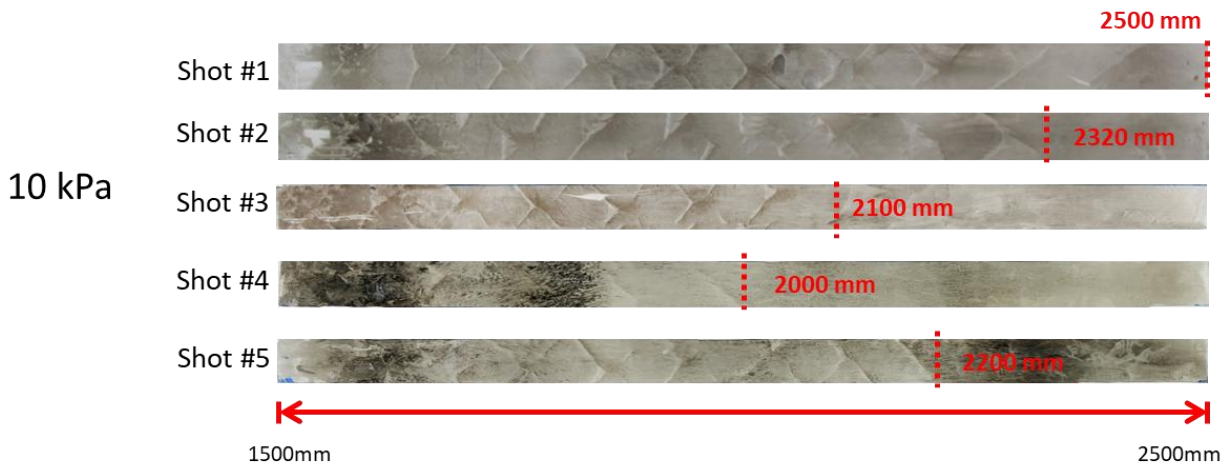
Fig. 2 Smoked foils obtained from the repeatability shots at the same condition
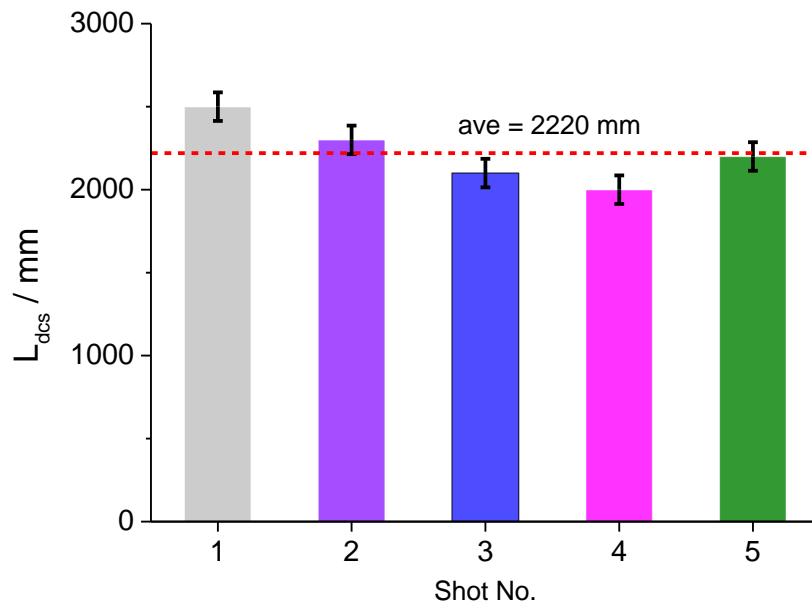(D=25 mm, p_0=10 kPa)

Fig.3 Experimental uncertainty of L_{dcs}

In addition, the uncertainty of the velocity should be considered. The response time (output rise or fall) of the fiber optic detector (IF-95OC) is 0.1 μs. Since the distance between each optical fiber is 10 cm, if it is a CJ detonation (e.g., V_{CJ}=2280 m/s at p_0= 8 kPa), the interval time between two adjacent fibers is 43.86 μs, if the response time of the optics is considered, the error of the velocity measurement is 0.44%.

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3. Results and discussions

3.1 Maximum length of detonation cellular structure (L_{dcs})

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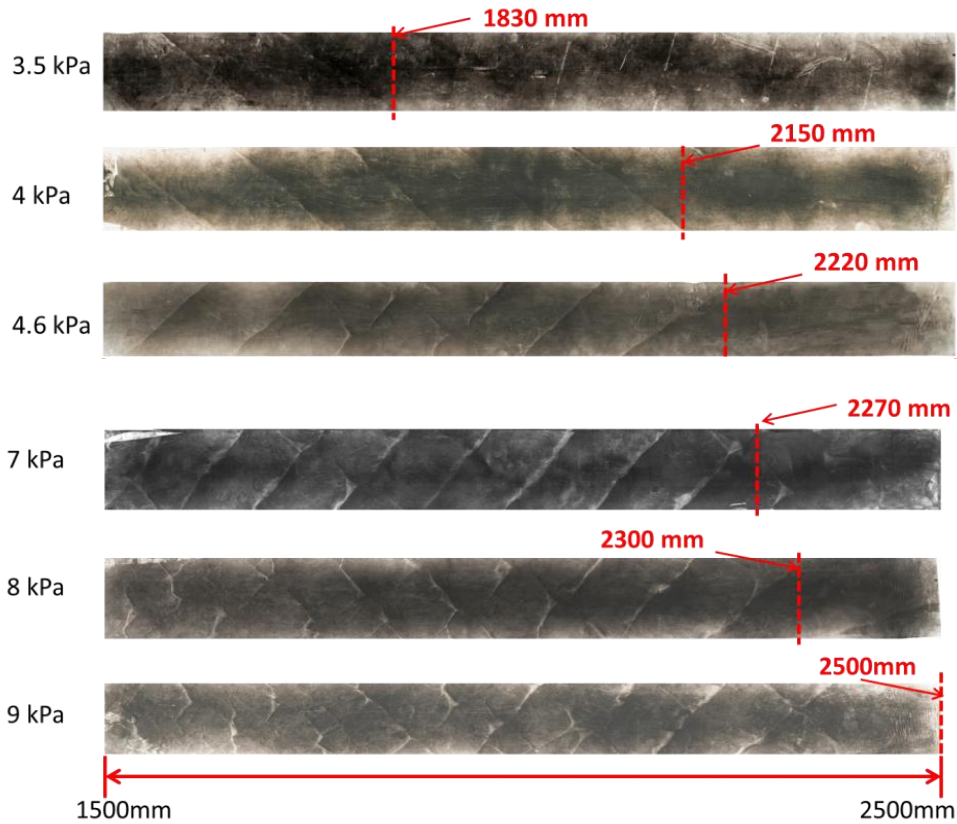
Fig. 4 Detonation cellular pattern in the $D = 36$ mm tube with the variation of p_0

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Figure 4 shows the results of L_{dcs} for the $CH_4 + 2O_2$ mixture in a $D = 36$ mm circular tube under various initial pressures. At $p_0 = 3.5$ kPa, the detonation structure first appears as a single-headed spin, which is the typical behavior as the detonation is approaching its limit. For this case, $L_{dcs} = 1830$ mm, after which no cellular structure can be found. The latter indicates that the precursor shock decouples from the following reaction zone and hence, the failure of detonation occurs. After $L_{dcs} = 1830$ mm, there are still some weak helical traces that appear seemingly, those are not representative of detonation structure, but only faint pressure waves pattern that reflected from the tube wall or soot displaced by the flame.

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For initial pressures below 9 kPa, cellular patterns can be observed only in part of the smoked foil, the propagation length of L_{dcs} is longer at higher initial pressure. At $p_0 = 9$ kPa, cellular patterns are registered on the complete foil and therefore, the critical pressure (p_c) for the 36-mm circular tube is accordingly 9 kPa. As p_0 increases higher than 9 kPa, it can be speculated that the detonation can be self-sustained in the current tube.

(a)

(b)
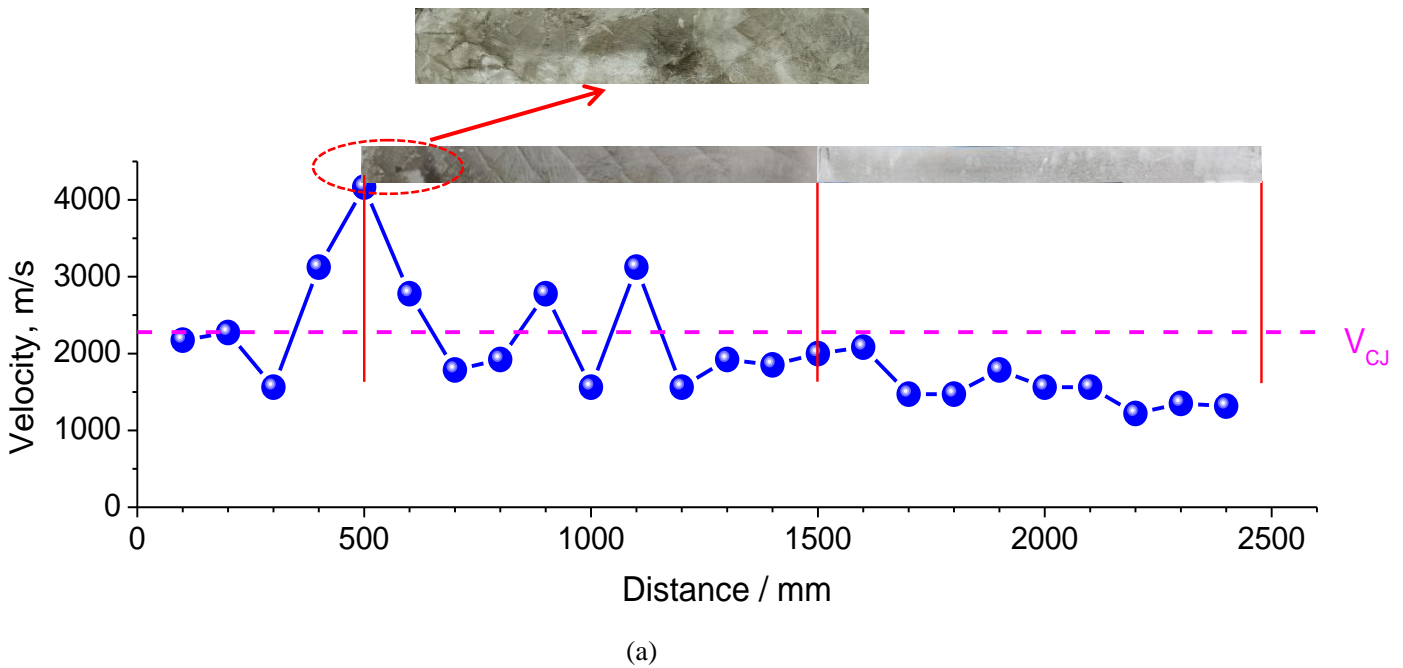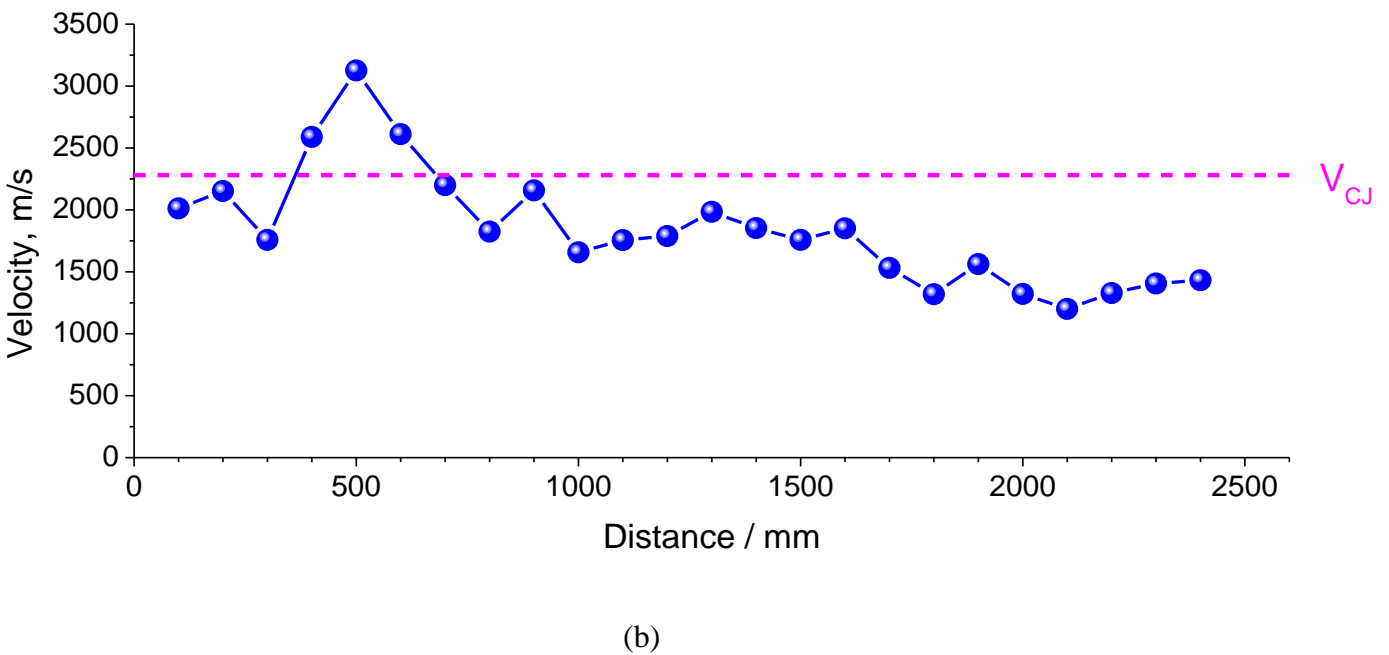
Fig.5 a) Local velocity and cellular pattern of detonation propagation in $D = 25$ mm tube ($p_0 = 8$

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kPa); and b) Local velocity of detonation propagation in $D = 20$ mm tube ($p_0 = 9$ kPa)

Local combustion wave velocities were simultaneously acquired together with the smoked foils. The local velocity results are used to further verify the propagation length L_{dcs} and failure of the detonation. Figure 5 (a) gives the detonation local velocity and the detonation cellular pattern in the $D = 25$ mm circular tube. The length of the smoked foil is 2000 mm, i.e., from $L = 500$~2500 mm. Figure 5 (a) indicates that, as detonation transmits into the 25-mm circular tube, the detonation first accelerates to an over-driven detonation ($L = 500$ mm), and the detonation front consists of multi-scale cells structure within the next 25 mm distance. As the detonation enters into a small diameter duct from a larger one, the precursor shock wave reflects from the wall, the shock wave is then strengthened and develops to a Mach reflection. If the reflection wave is sufficiently strong, a slightly over-driven detonation may be resulted but quickly relaxed. Similar local velocity evolution is also seen in Fig. 5 (b) for another tube size $D = 20$ mm tube at another initial pressure $p_0 = 9$ kPa.

The initially over-driven detonation can always be observed as boundary conditions when a detonation propagates into an abrupt area change, either as a detonation transmits through an obstacle, or propagates into a smaller tube from a larger one. For example, Wu and Lee [35] observed the detonation in $CH_4 + 2O_2$ is over-driven as it passes through an obstacle. Since we investigated the detonation propagation behavior in different diameter tubes, and those tubes' inner diameters are smaller than the test tube, hence the over-driven detonation is inevitable. For conditions well within the limits, the over-driven detonation quickly develops back to a self-sustained detonation propagating at about CJ detonation velocity. In the contrary, as the conditions outside the limits, the initially over-driven detonation relaxes rapidly within few diameter tubes and the detonation wave continues to decay, and subsequently fail to a fast flame,

see Fig. 5. Therefore, the appearance of an initially over-driven detonation does not appear to affect the propagation behavior in the smaller tubes and that the detonation propagation behavior is mainly governed by the boundary conditions and the thermodynamics of mixtures in the small tubes.

Figure 5 (a) also shows after a short, early transient appearance of an over-driven detonation, the detonation structure changes quickly to a single-headed spin. As the detonation continues to decay and the combustion wave's propagation velocity is significantly below the CJ value, the spinning structure disappears from the foils, which indicates the detonation attenuates into a fast deflagration wave with low velocity. Therefore, the propagation velocity behavior agrees well with the cellular pattern evolution. Although a fluctuation of velocity is observed from $L = 900$ to 1100 mm, this velocity fluctuation is totally different from the galloping propagation mode. For a galloping detonation, the wave first decays to about half CJ detonation velocity from an overdriven state, and the cellular pattern disappears. Subsequently, this low velocity combustion wave accelerates once again back to the overdriven state, and the cellular structure recovers. The velocity of a galloping detonation fluctuates from 1.5 V_{CJ} to 0.4 V_{CJ}. The period of one single cycle of galloping detonation is about 350 times of tube diameter. In this study, although similar velocity fluctuation is observed ($L = 900$ to 1100 mm), the cycle is much less than the typical galloping detonation. Furthermore, it can be seen from the smoked foil that, the single-headed spinning structure is continuous from $L = 500$mm to 1500 mm. Therefore, for the distance of $L = 900$ to 1100 mm, it is only some local fluctuation of the propagating detonation, which is also a typical propagation mode near the limits for an unstable mixture like methane-oxygen. Figure 5 shows that, the velocity at $L = 100$ mm is approximately equal to the CJ detonation value of CH_4-2O_2 (2280 m/s).

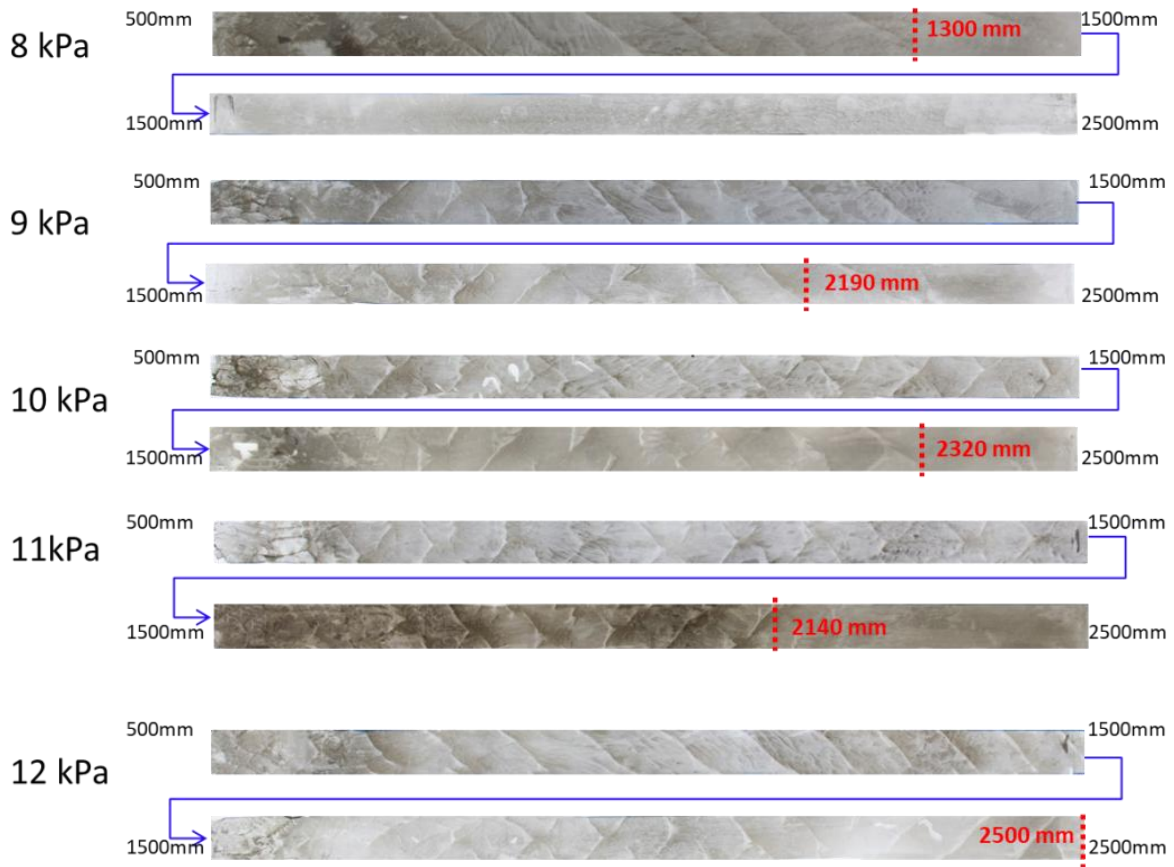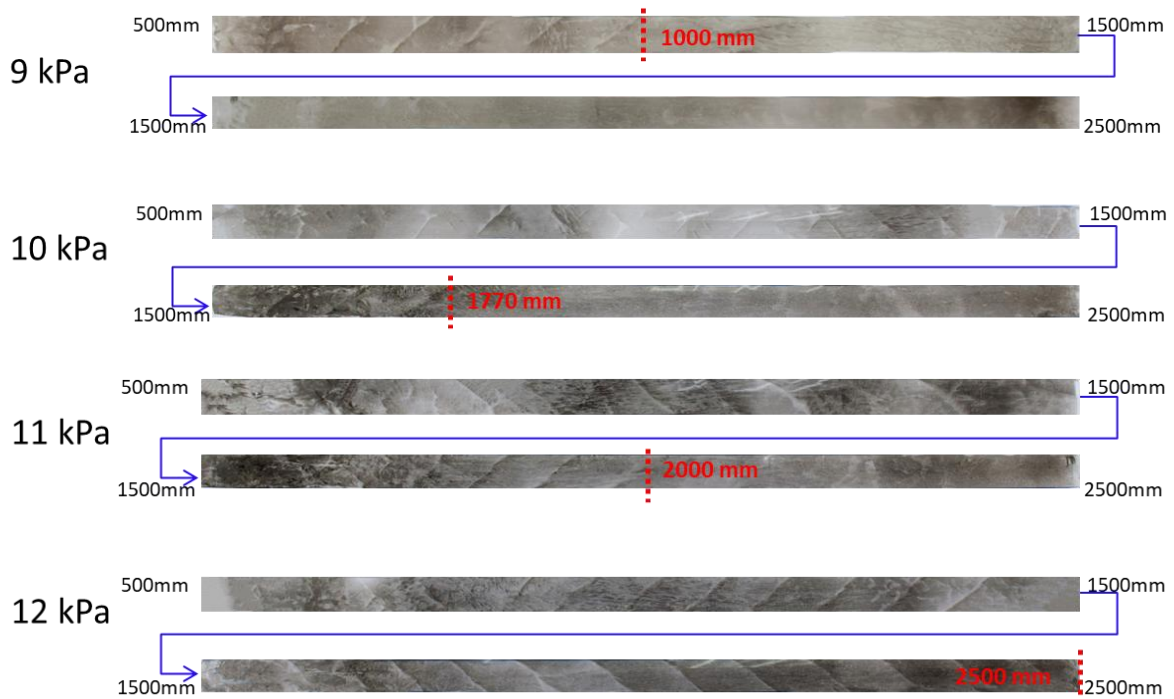Fig.6 Detonation structure with the variation of p_0 (D = 25 mm)

Figure 6 gives the detonation cellular pattern in the D = 25 mm circular tube for increasing initial pressure from 8 to 12 kPa. At p_0 = 9 kPa, the single-headed spinning structure maintains to L= 2190 mm. In general, with an increase of p_0, L_{dcs} is consequently elongated. Because of the experimental uncertainties, fluctuation of L_{dcs} can be observed, e.g., as in the p_0 = 11 kPa case. As p_0 increases to 12 kPa, the detonation propagates till the end of the tube (L = 2500 mm) without failure. For D = 25 mm circular tube, the critical pressure is thus p_c = 12 kPa. In some cases, the value of L_{dcs} at higher initial pressure is smaller than that in lower initial pressure, e.g., L_{dcs} in p_0 = 11 kPa is smaller than the result in p_0 = 10 kPa. This variation is a common phenomenon for detonation experiment, especially for unstable mixtures. For these mixtures, detonation initiation is characterized by local explosion; the occurrence of the local explosion has a certain randomness due

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Figure 7 shows the detonation propagation behavior in the $D = 20$ mm tube. $p_0 = 9$ kPa is the initial pressure at which some cellular structure in the 2000-mm long smoked foil can still be observed. As p_0 increases to 10 kPa and 11 kPa, L_{dcs} increases to 1770 mm and 2000 mm, respectively. At $p_0 = 12$ kPa, a robust single-headed spinning structure throughout the whole 2000-mm long smoked foil can be found.

Fig.7 Detonation structure with the variation of p_0 ($D = 20$ mm)

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Figure 8 gives the cellular pattern in the smallest tube considered in this work, i.e., $D = 13$ mm.

At $p_0 = 10$ kPa, the detonation fails after $L = 680$ mm ($L_{dcs}/D = 52.3$). For the rest of the foil ($L =$

680 mm ~ 2500 mm), there is no noticeable structure. At $p_0 = 11$ kPa, L_{dcs} extends to 1700 mm

($L_{dcs}/D = 130.8$). At $p_0 = 12$ kPa, detonation failure occurs at $L = 830$ mm. It is interesting to note

that at $L = 2040$ mm, detonation re-initiation occurs. After the re-initiation, the detonation is

over-driven characterized by small fine-scale cells pattern. Subsequently, the detonation decays to

the CJ state with normal cellular structure near the end of tube. Later, the cellular pattern turns into

spinning again. This observed phenomenon agrees with the galloping detonation that is reported in

previous literatures [31, 37, 57, 58].

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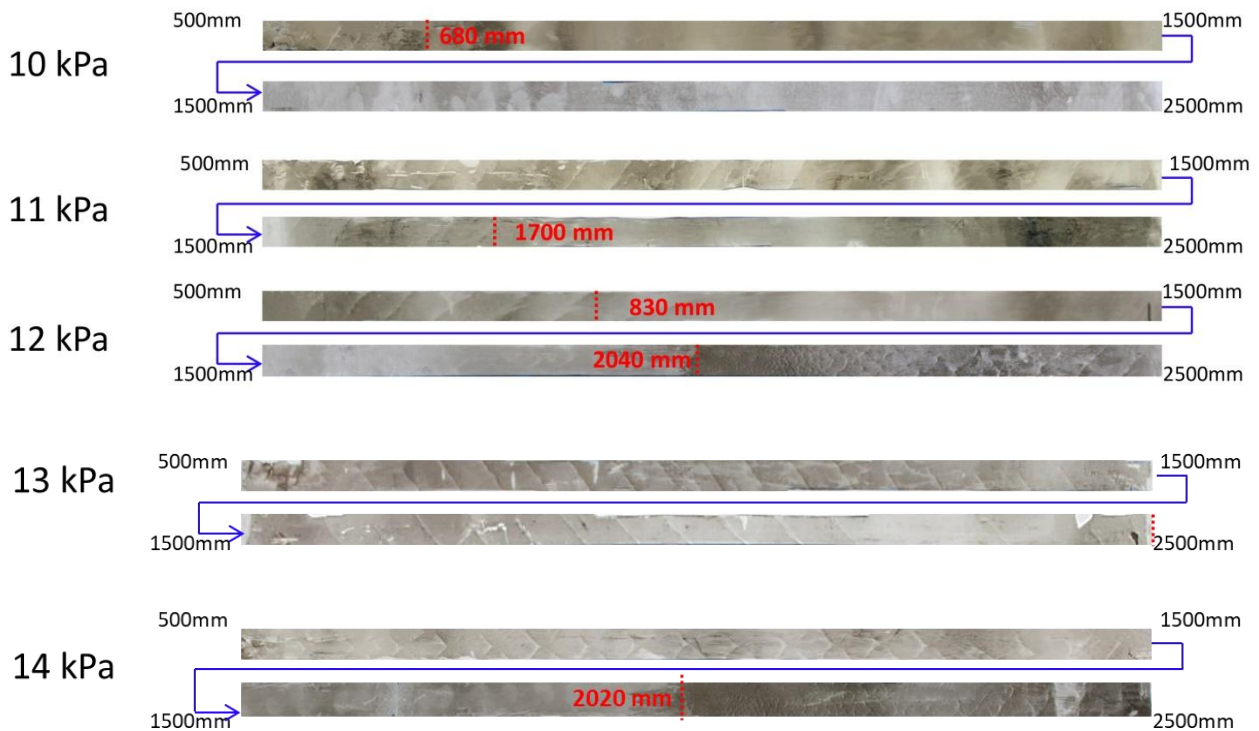
Fig.8 Detonation structure with the variation of p_0 ($D = 13$ mm)

It is well known that for a detonation propagating in the galloping mode, the wave first decays to a low-velocity regime from an overdriven state, and later it accelerates back to the overdriven detonation. One cycle period of a single galloping detonation usually needs a length of several hundred of tube inner diameter. At $p_0 = 12$ kPa, the distance from the failure to re-initiation is 1210 mm, $L/D = 93$. As p_0 is slightly increased to 13 kPa, only single-headed spinning structure from $L = 500$ mm to $L = 2500$ mm is observed. At $p_0 = 14$ kPa, galloping detonation with failure and re-initiation behavior can be found, this phenomenon is similar as in the $p_0 = 12$ kPa case, but with a shorter run-up length. When the initial pressure further increases to 15 kPa, 16 kPa and even 18 kPa, an entire foil with cellular structure can be observed. Hence, for the $D = 13$mm tube, $p_c = 14$ kPa.

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3.2 Scaling analysis of detonation failure behavior

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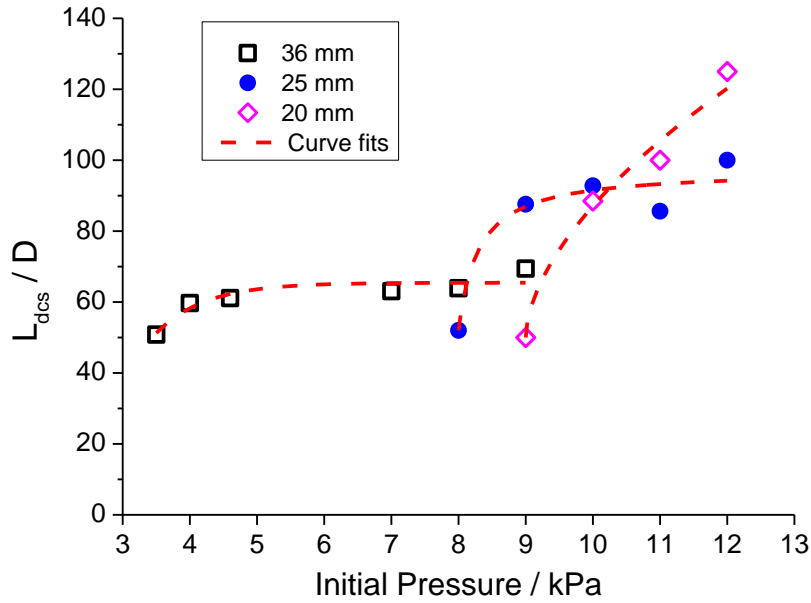
Fig. 9 L_{dcs}/D as a function of p_0 in tubes

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As a detonation propagates in tubes, besides the thermodynamic properties of the mixture, the

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propagation behavior is greatly dependent on the boundary condition. The relationship between the

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detonation propagation behavior and the initial pressure in different diameter tubes is given by Fig.

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9. For the $D = 13$ mm case, galloping behavior is observed, i.e., after the disappearance of the

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spinning detonation, the deflagration can eventually re-initiate and form an over-driven detonation

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after a certain distance. This criterion that is used to estimate the L_{dcs} in larger diameter is thus not

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suitable for the $D = 13$ mm case and hence, only $D = 36$ mm, 25 mm and 20 mm results are

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considered for discussion in Fig. 9. For the $D = 36$ mm circular tube, as p_0 increases from 3.5 kPa to

404

9 kPa, the value of the ratio between L_{dcs} and tube inner diameter (D) slowly goes up. In other

405

words, the detonation propagation is not sensitive to p_0 in the larger inner diameter tube ($D = 36$

406

mm). In the middle-size diameter circular tube, i.e., $D = 25$ mm, it is clear that L_{dcs}/D increases with

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the increase of initial pressure, which indicates a small variation of the initial pressure may cause a

408

409 long distance for either successful propagation or failure of a detonation. Finally, for the $D = 20$ mm

410 diameter tube, the value of L_{dcs}/D increases very abruptly with increasing initial pressure, indicating

411 L_{dcs}/D has a strong dependence on its p_0.

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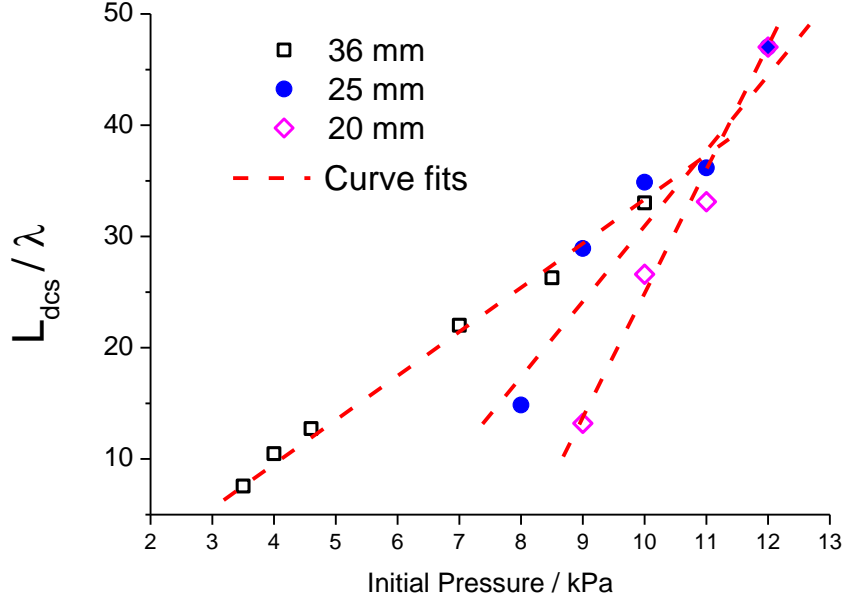
Fig. 10 Variation of L_{dcs}/λ with p_0

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416   As the limits are gradually approaching, the unstable mode of detonation turns into the lowest

417 mode of acoustic wave. This acoustic mode is dominated by the tube's characteristic dimension.

418 Therefore, the characteristic length scale of the cellular detonation front is correlated with the

419 characteristic length scale of the tube at the limit condition. In this work, an attempt is made to

420 correlate the two important length scales near the detonation limits used in this work, i.e.,

421 detonation cell size λ and L_{dcs} as proposed in this work. Figure 10 gives the variation of L_{dcs}/λ with

422 the initial pressure for different tubes. In this work, the detonation cell sizes data are taken from

423 Zhang et al. [41]. The relation between cell size and p_0 is given by the power function as follows:

424 $$\lambda = 1127 \cdot p_0 \ [\text{kPa}]^{-1.229} \tag{1}$$

425   In Fig. 10, it is found that a linear relationship between L_{dcs}/λ and p_0 for a specific tube can be

426   obtained. The increase of L_{dcs}/λ is however more abrupt in the smaller diameter tube, which

427   confirms again the propagation of detonation is more dependent on the initial pressure in smaller

428   diameter tube.

429   Another noteworthy observation from Fig. 10 is that for different diameter tubes, the minimum

430   failing length L_{dcs} observed at low initial pressures is of the order of 5 to 14 λ. The upper value, i.e.,

431   is observed for smaller tube diameter D of which the boundary layer effect not only promotes the

432   failure, but is found to sustain the wave propagation through flow fluctuations at lead shock

433   interacting with the boundary. For reference, this minimum L_{dcs} range is larger as compared to the

434   failure length of 3~7 λ reported in Radulescu and Lee [53] for the distance traveled by the

435   attenuated detonation in porous wall tubes before it fails. The difference can be explained as follows.

436   One should notice that the failure length measured in Radulescu and Lee [53] is defined as the

437   distance between the locations where the first expansion wave reached the axis to the point where

438   the detonation fails, while in the present study, it is defined from the beginning of the test tube

439   section. In addition, the failure mechanism of the present problem and that Radulescu and Lee [53]

440   are also different. The failure mechanism in Radulescu and Lee is attributed to the losses and mass

441   divergence into the porous walls, however, in this study, the detonation failure is mainly caused by

442   tube scale or geometry influencing the effects of boundary layer. Finally, the cell size measurement

443   and correlation for low pressure conditions generally have a significant uncertainty and this alone

444   already leads to some discrepancy in the scaling. Although there exist some similarities between the

445   present work and that of Radulescu and Lee [53] and a similar failure length has been defined, it is

446   not an attempt here to conclude any quantitative agreement between the two works due to the

447   aforementioned differences inherent in both phenomena.

448   With decreasing p_0 and D, the boundary layer displacement thickness increases accordingly

449   [34]. The enlarged boundary layer displacement thickness in the smaller scale tube causes more

450   momentum losses through the flow divergence, and faster distribution of curvature over the whole

451   detonation front, which eventually results in an earlier failure of the detonation propagation.

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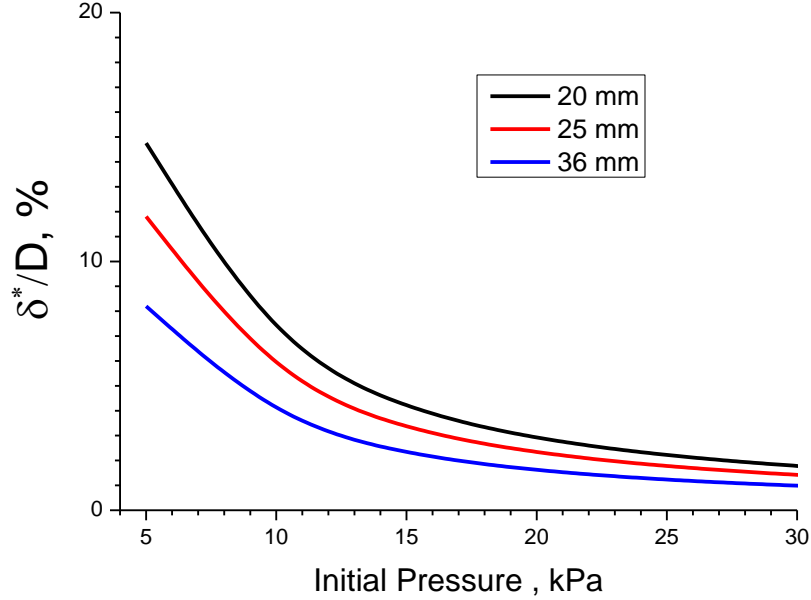
Fig. 11 Variation of δ^* with p_0 in different tubes

455

456   The boundary layer displacement thickness (δ^*) as a function of p_0 in $D = 36$ mm, 25 mm and

457   20 mm tubes are given in Fig.11. δ^* was given by Gooderum [59], which is as follows:

$$\delta^* = 0.22x^{0.8}\left(\frac{\mu_e}{\rho_0 V}\right)^{0.2} \tag{2}$$

459   in which, x represents the reaction zone thickness, Lee [20] and Gao et al. [34] argued $x = 1.5\lambda$. μ_e

460   refers to the viscosity, V is the detonation velocity and ρ_0 is the initial density. Figure 11 shows

461   that, with the decrease of initial pressure and the reduction of the tube diameter, the value of δ^*

462   evidently increases.

463   For conditions at which δ^*/D is small, either increasing D or p_0, the distribution of global

464   curvature due to the presence of boundary layer becomes less significant and the failure is thought

to be more dominant by the ability to maintain half a detonation cell in the tube. In other words, the failing length should then be governed primarily by the rate of cell decay or transverse waves attenuation, i.e., how fast cells evolve or disappear. This is because as detonation suffers more losses from the wall, the losses to the tube walls are communicated with the entire front more rapidly, and the failure of detonation occurs more promptly. For $D = 25$ and 36 mm, it is interested to note in Fig. 9 that the two curves tend to plateau as pressure increases or the boundary layer thickness relative to the tube diameter decreases.

It is important to emphasize that the failure length scaling investigated here is valid for conditions "far" below the limit. In fact, despite some criteria are proposed in the literature, it remains ambiguous to define exactly the critical detonation limit value due to different unstable behaviors observed. Due to the inherent unstable behavior of near-limit detonation propagation, it would be difficult to validate the similar scaling at the proximity to the failure limit. Nevertheless, we conjecture that there should be a finite failure length at the limit. For the case of unstable detonations, if the limit phenomenon can be attributed to the ability to maintain half a detonation cell, the failure length should be corresponding to the inherent condition when a finite cell size cannot be accommodated. Thus, one cannot determine an increasing failure length toward infinitely by reducing fractionally a detonation cell as it approaches to the critical limit. Furthermore, the methane-oxygen mixture considered here is unstable and the cellular front pattern is irregular. The presence of local instability effects could also affect the scaling at the proximity to the failure limit. In fact the inherent instabilities are ingredient of different unstable behavior for near-limit detonation propagation.

Figure 11 also shows that at low pressure, the boundary layer thickness growth is comparable to the tube diameter and engulfs the whole reaction structure. The failure length should be expected

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488 to scale with D. This is indeed observed in Fig. 9. The L_{dcs}/D of all tube diameters is approximately

489 50 for their lowest initial pressure, at which the boundary layer displacement thickness δ^* is

490 comparable to the tube diameter D. At these conditions, the curvature effect due to the lateral mass

491 divergence can fail the detonation at a shorter length L_{dcs} for smaller tube due to the shorter time it

492 takes for the global curvature to distribute over the whole front [53, 60].

493　　Similar to Figs. 9 and 10, the relationship between ratio of L_{dcs}/δ^* and the initial pressure is

494 given in Fig. 12. The slope of L_{dcs}/δ^* is more steep in the smaller diameter tube, this result

495 demonstrates again that the boundary layer displacement thickness in the smaller diameter tube

496 greatly affects the earlier extinction of a detonation.

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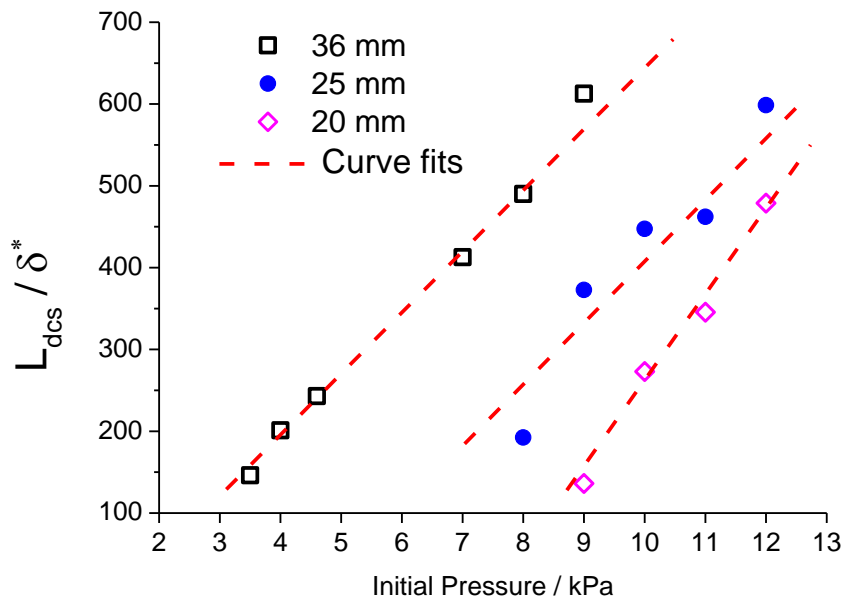
498

499 Fig. 12 Variation of L_{dcs}/δ^* with p_0 in different tubes

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3.3 Determination of detonation limits

Table. 3 Parameters at the critical condition of detonation limits

D/mm	p_c / kPa	λ/mm	D/λ
36	9	75.71	0.48
25	12	53.16	0.47
20	12	53.16	0.38
13	15	40.41	0.32

Table 3 shows the results of the critical pressure (p_c), above which a detonation can self-sustain throughout the entire tube defined by the existence of continuous cellular detonation structure on the whole foil. λ at the critical pressure is also scaled with its tube inner diameter. For circular tubes, Lee [20, 61] proposed $\lambda = \pi \cdot D$ as a limit criterion, where λ represents the transverse wave spacing or cell size, and D is the tube diameter, this criterion corresponds to the first onset of single-head spin. When the structure of detonation is single-headed, the characteristic dimension is the circumference of the tube, i.e., $\pi \cdot D$. This criterion was verified by other researchers, e.g., Dupré et al. [62], Yoshida et al. [36], Fischer et al. [63] and Gao et al. [30]. As can be seen from Table 3, $D/\lambda \sim 0.41$, which approaches well to the detonation limit criterion $\lambda = \pi \cdot D$ within the experimental uncertainties. Generally λ is inversely proportional to the initial pressure, i.e., $\lambda \sim p_0^{-1}$. Given the scaling $\lambda = a \cdot D$ where the parameter $a > 1$, it indirectly demonstrates that the limit is more sensitive to p_0 governing the detonation structure than the effect of tube scale.

In summary, some remarks can be made with regards to the effects of p_0 on the detonation limit phenomenon. It is perhaps well established that p_0 affects the dynamic detonation parameters. In this study, the test mixture is CH_4-$2O_2$, which is considered an unstable mixture (i.e., with irregular cellular pattern). For unstable mixtures, the detonation propagation limit criterion $\lambda = \pi \cdot D$ holds in macro-scale ducts. Yoshida et al. [36] proposed a factor (α) that determines the detonation

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limit, i.e., $\alpha = \pi \cdot D / \lambda$. When α equals to 1, the detonation limit is approached. If $\alpha < 1$, the single-headed spinning structure is observed and afterwards this structure disappears eventually, which indicates detonation failure. It is well observed that with the decreasing of p_0, the detonation cell size λ increases exponentially, which renders the value of α much smaller than 1, and the detonation failure occurs. The ability of detonation propagation in tubes thus depends on the ability of the mixture to generate cells or transverse waves within, of which the initial pressure is one of the governing parameter.

From a purely thermodynamic and chemical kinetic point-of-view, although it could be a secondary effect, decreasing initial pressure reduces the detonation strength and elongates the induction zone of the detonation structure. The latter also explains why the cell size decreases as initial pressure increases. For a fixed diameter tube, it leads the detonation structure more susceptible to losses making it more prompt to failure when the boundary layer thickness becomes comparable to the reaction zone thickness. The initial pressure also affects the boundary layer displacement thickness δ^*. The δ^*/D appears to be a significant parameter to explain different trends shown in Fig. 9 and Fig. 10. The initial pressure, which governs the value of δ^*/D, could therefore dictate whether the failure mechanism is primarily dominated by the lateral mass divergence resulting a global frontal curvature or the rate of cells attenuation.

4. Conclusions

In this study, the dynamic behavior of detonation propagation and failure at the limits was investigated experimentally. Simultaneous velocity measurement and smoked foils were used to observe velocity deficits and the evolution of the detonation cellular structure, from which limits (i.e., critical pressures) were defined. An alternate characteristic length (L_{dcs}), defined as the length

from the start of the test tube section to the location where no cellular detonation structure, is recorded with varying initial pressure in four different inner diameter tubes, i.e., D = 36, 25, 20 and 13 mm. The quantitative relation between the cellular detonation propagation distance, the thermodynamic properties of the mixture, and the tube geometry were explored. The results show that L_{dcs} generally decreases with decreasing initial mixture pressure, and it decreases faster in smaller diameter tubes. In this study, the detonation galloping mode is only observed in the D = 13 mm diameter tube. With the increase of p_0, the length of the galloping cycle is reduced. By scaling L_{dcs} with tube inner diameter (D) and detonation cell size (λ), it is found that the decrease of L_{dcs}/D and L_{dcs}/λ are more abrupt in smaller diameter tubes with decreasing initial pressure. It thus suggests that the detonation propagation dynamics is more sensitive to the initial pressure in the smaller diameter tube. The latter is explained according to the argument of the significant boundary layer displacement thickness growth at low initial pressure and the curvature due to the lateral mass divergence. The distribution rate of global curvature over the whole detonation front due to the boundary layer effect is faster in smaller tube and thus it leads to a more abrupt decrease sensitive to initial pressure.

For increasing pressure closer to the failure limit from below, the boundary layer displacement thickness is becoming less comparable to the tube diameter. The failing length no longer scales with D based on the global curvature. The failure mechanism can be thought to be more dominant by the rate of transverse wave attenuation or cell disappearance. Lastly, by analyzing the critical condition of detonation limits in different tubes, once again it confirms that $\lambda = \pi \cdot D$ provides an appropriate limit criterion for detonation propagation in accordance with the literature.

Nomenclature	
CJ	Chapman-Jouguet
V_{CJ}	CJ detonation velocity
D	Tube inner diameter
p_c	Critical pressure for detonation limits
p_0	Initial pressure
T_0	Initial temperature
λ	Detonation cell size
δ^*	Boundary layer displacement thickness
L	Length of tube
L_{dcs}	Maximum length of detonation cellular structure
AC	Annular channel
CT	Circular tube
H	Height of channel
χ	Stability parameter
φ	Equivalence ratio

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