Detonation limits in highly argon diluted acetylene-oxygen mixtures

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1. Introduction

Detonation limits, the conditions outside of which a self-sustained detonation wave can no longer propagate, is an important fundamental as well as practical problem. The development of a theory for predicting the limits as well as the experimental measurement of the limits are challenging due to the strong influence of boundaries which introduce not only losses of heat, momentum and mass [1], but can also intervene with the inherent instability of the detonation front [2]. Because of all these effects, the boundary between failure and successful propagation is not precise, and a spectrum of unstable near-limit phenomena (e.g. spinning, galloping, pulsating and stuttering modes) can occur. The study of the propagation limits behavior in narrow gaps (e.g., thin annular channel) is important from the aspect of safety assessment for fuel/oxygen mixtures, because the maximum safe gap determined from experiment is an important parameter for deflagration or detonation hazard assessments.

It was previously suggested that the limits in stable mixtures (e.g., highly argon diluted acetylence-oxygen) can be determined due to the absence of marginal behavior of the detonation wave near the limits that can lead to large velocity fluctuation typically observed in unstable mixtures without any argon dilution. In the stable mixtures, it was suggested that detonations characterized by a low degree of instability (i.e., with regular cellular structures and weak transverse waves) are usually less robust than unstable detonations with irregular and unstable structure, this is because stable mixture is less prompt to form the localized explosions and has the low ability of facilitating the initiation or the onset of a detonation than unstable mixture [3]. Therefore, in highly argon diluted mixtures, cellular instabilities do not play a prominent role on the propagation of a stable detonation. The detonation limits are governed solely by losses, and therefore one can isolate the propagation mechanism of a stable detonation to investigate the effect of boundary conditions on the limits [4]. Those marginal phenomena observed in unstable detonation are also absent in stable detonation, making it possible to define clearly the limits.

In this study, the velocity deficits and the detonation limits in stoichiometric acetylene-oxygen mixtures highly diluted with argon in two different geometries (i.e., a small circular tube and three narrow annular channels) are reported and compared. For the highly argon diluted mixtures tested in this investigation, the detonations are considered as relatively stable. The mechanism causing the limit is proposed to be due to boundary layer effects at the wall and consequently, the flow divergence in the reaction zone gives rise to a critical frontal curvature causing detonation failure. Previous literatures [5-7] have confirmed that the mass flow divergence results in an axial momentum loss, resulting in the velocity deficit.

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According to the aforementioned failure mechanism for highly argon dilution mixtures, it is suggested that there should be a simple scaling between the critical diameter for the circular tube and hydraulic diameters for annular channels with the characteristic length scale of the detonation structure for the detonation limits. The systematically scaling analysis is conducted to explore the general feature of the detonation near-limit behavior in stable mixtures.

2. Experimental details

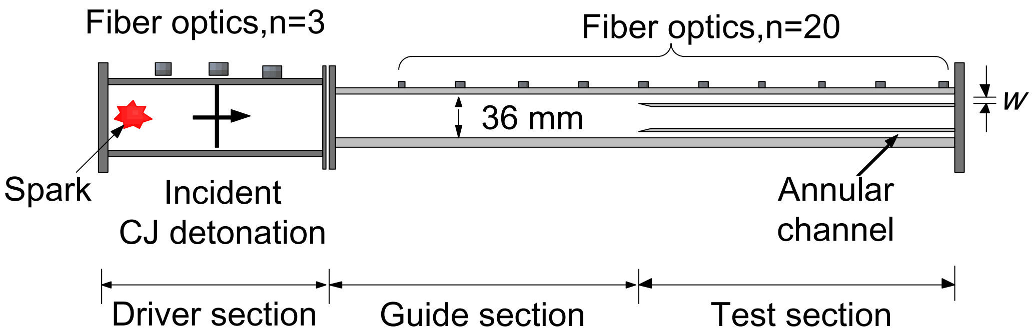

Figure 1. Schematic of the experimental apparatus

A circular tube and three annular channels were used in the present investigation. A schematic of the experimental setup is shown in Fig. 1. A 1.2-m long steel tube with an inner diameter of 68 mm, filled with a more sensitive mixture (i.e., equi-molar acetylene and oxygen, C_2H_2-O_2), was used as a driver in order to form a stable Chapman-Jouguet (CJ) detonation in the guide section prior to the detonation wave entering the test section. In the experiment, there is a diaphragm locates between the driver and guide section to separate the driver and test mixtures. To minimize the influence of the driver section on the results, we only focused on the velocity in the test section, since the test section is far enough to the driver section. For the annular channels experiments, the narrow channel section was created by inserting a smaller inner diameter tube into a 2.5-m long tube with an inner diameter of 36 mm. Three different channel heights were considered in this study, i.e., w = 2, 4.5 and 7 mm. With the large aspect ratio, the annular channels thus mimic a continuous "quasi two-dimensional" rectangular channel [8]. The same 2.5-m long, 36-mm diameter tube is used also to determine the detonation limits in a circular geometry.

Fiber optics of 2.2 mm in diameter were spaced periodically along the entire length of the test section and guide section to measure the time-of-arrival of the detonation at various optical probe locations. At least three shots were repeated for each condition. When the detonation is well within the limits, the maximum difference of the velocity measurement is ~10 %. When the detonation approaches its limits, the repeatability of the measurement is poor, and large fluctuation can be observed.

In this study, stoichiometric mixtures of acetylene-oxygen with 70 % and 85 % (vol.) argon dilution were tested. The explosive mixture was prepared beforehand in separate gas bottles by the method of partial pressure. The initial pressure of the test mixture was monitored by an accurate digital manometer model OMEGA PX409-150A5V (0-150 PSI) with an accuracy of ± 0.08 % full scale. The gases were allowed to mix in the vessel for at least 24 hours in order to ensure homogeneity. For any given experiment, the detonation tube was evacuated to lower than 10 Pa. The entire apparatus was then filled from both ends to the desired initial pressure. The sensitivity of the explosive mixture was varied by changing the initial pressure, p_0.

3 Results and discussion

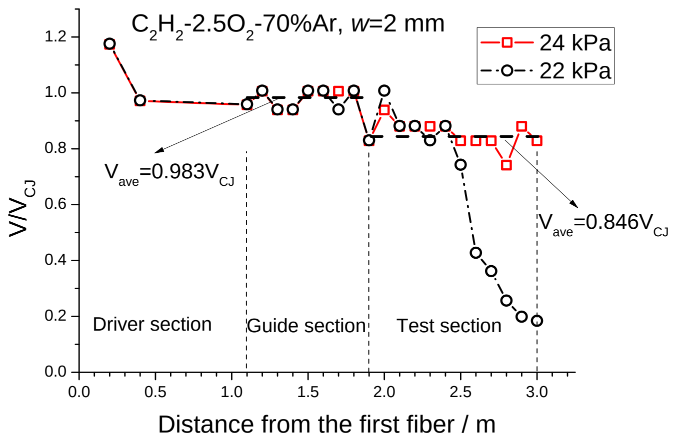

Fig.2 Normalized velocity at the conditions above and below the limit pressure

Figure 2 shows a typical velocity plot in different sections of the test apparatus for the C_2H_2-2.5O_2-70%Ar mixture at initial pressure above and below the limit. The average detonation velocity in the guide section for both sets of data is close to the theoretical CJ value, i.e., V_{ave} = 0.983 V_{CJ}. As the detonation propagates into the annular channel (w = 2 mm) with an initial pressure higher than the limit value, i.e., p_0 = 24 kPa, detonation still propagates with a mean velocity value of 0.846 V_{CJ}. On the other hand, detonation velocity abruptly decays as the initial pressure decreases to p_0 = 22 kPa, which is due to the more loss from the wall that the detonation experiences.

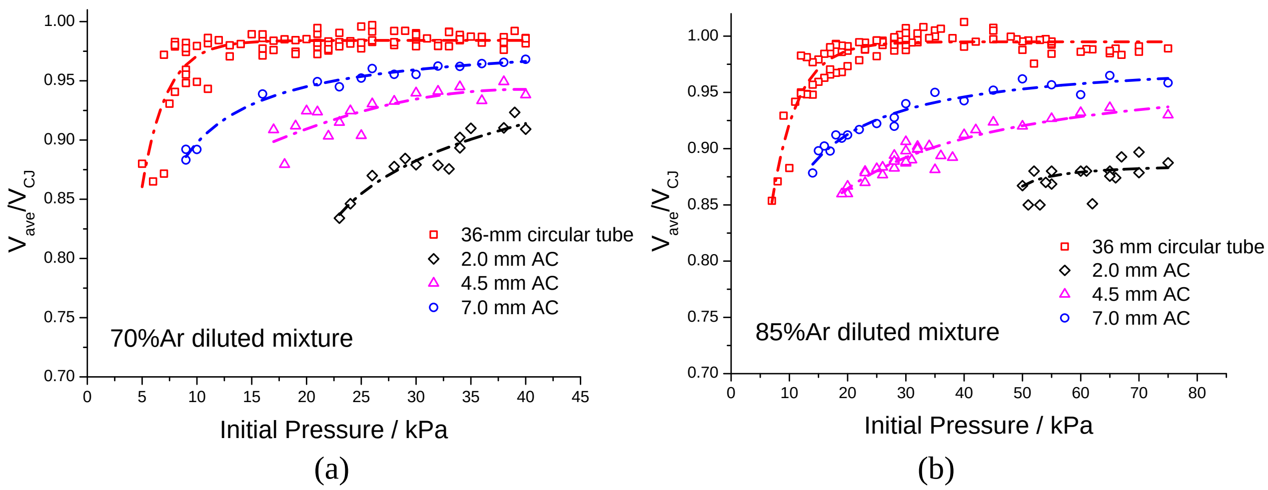

Fig.3 Normalized velocity as a function of initial pressure in circular tube and annular channels (AC) in a) 70%Ar and b) 85%Ar diluted mixtures (dash lines are the curve fits)

Figure 3 shows the variation of the average detonation velocity (V_{ave}) normalized with the CJ value as a function of initial pressure in the 36-mm circular tube and different annular channels for C_2H_2-2.5O_2-70%Ar and C_2H_2-2.5O_2-85%Ar mixture, respectively. The data in Fig. 3 are all the "steady" velocity measurement from the photodiode signals. The average velocity (V_{ave}) as a function of initial pressures (p_0) is plotted for different tube geometries (i.e., the circular tube and annular channels) obtained from the present experiments. As the limit is approached by reducing the initial pressure, the detonation velocity progressively decreases and deviates from the CJ value. In these velocity deficit plots, the value of critical pressure below which a steady detonation does not occur can be extracted from the abrupt decay in the velocity, thus defining the limits. The limit values and also the maximum velocity deficits obtained from Fig. 3 are summarized in Table 1. In Table 1, *D* and *w* represent the length scale of round tube and annular channel. p_c is the critical limiting pressure, below which the detonation fails. λ and Δ_I are the detonation cell size and induction zone length at the critical pressure. Here, Δ_I is determined as the length of the thermally neutral period in detonation structure. It is defined as the distance from the leading shock to the maximum temperature gradient in the ZND temperature profile as used in previous studies [9, 10].

Table 1 Summary of the experimental results of highly argon diluted mixtures

Mixtures	*D* or *w* / mm	p_c / kPa	Velocity deficit / %	λ / mm	D_H/λ	Δ_I / mm	D_H/Δ_I
C_2H_2-2.5O_2-70%Ar	36	5	13.5	40.5	0.89	1.638	21.98
	7	9	11.4	18.5	0.76	0.845	16.57
	4.5	16	12.1	8.6	1.05	0.442	20.36
	2	23	16.6	5.3	0.76	0.294	13.62
C_2H_2-2.5O_2-85%Ar	36	7	14.6	72.9	0.49	2.677	13.45
	7	14	12.2	32.7	0.43	1.244	11.26
	4.5	19	13.9	23.0	0.39	0.887	10.14
	2	50	15.0	7.5	0.53	0.304	13.15

An interesting phenomenon can be observed from Fig. 3 and Table 1 that, the mean maximum velocity deficit is about 14 % of V_{CJ} for those stable mixtures, and this value is consistent in both the 36-mm inner diameter circular tube and the annular channel gap with different scales (i.e., w = 2, 4.5 and 7 mm). This maximum velocity deficit is very close to the result from Wu and Lee [11], in which they reported a velocity deficit of 18 % of V_{CJ} for the same two stable mixtures. The velocity deficit limit of detonation in stable mixtures is different from the unstable mixtures in annular channels. For example, methane-oxygen mixture is commonly considered as an unstable mixture, Zhang et al. [12] reported the velocity deficits in methane-oxygen mixtures with three different fuel compositions (i.e., CH_4-1.5O_2, CH_4-2O_2, CH_4-4O_2) in the same channel gaps, they found the velocity deficit increases as the channel gap is reduced for those mixtures. Lee et al. [13] reported the similar velocity deficit behavior using different scale channel gap for stoichiometric methane-oxygen mixture. In the study of Lee et al. [13], they also studied another unstable mixture, i.e., C_2H_2-5N_2O-50%Ar, it was confirmed that the velocity deficit increases from 17 % to 27 % as the channel gap is reduced from 9.525 mm to 3.175 mm.

A least-square regression of these data gives the power-law best fit correlation between cell size (acetylene-oxygen with 70% and 85% argon dilution) and initial pressure as:

$$\lambda[\text{mm}] = a \cdot \left(p_o[\text{kPa}] \right)^b \begin{cases} a = 348.08 \; b = -1.336; \; 70\%\text{Ar} \\ a = 691.32 \; b = -1.156; \; 85\%\text{Ar} \end{cases} \quad (1)$$

where the cell size, λ, is the length scale that characterizes the sensitivity of the explosive mixture. Here, we introduce hydraulic diameter (D_H) to compare all the experimental data based on a standard criterion. D_H is a commonly used term when handling flow in noncircular tubes and channels, which was also applied in our previous study [8] to investigate the prediction model of detonation limit. Using this term one can compare the results in circular tube and annulus at the same standard. For the annular channel, D_H is defined as:

$$D_H = 4 \times \frac{\pi(D^2 - d^2)}{4} \; / \; \left\{ \pi \left(D \; + d \; \right) \right\} = 2w \quad (2)$$

in which, D and d are the inner diameter of large tube and outer diameter of smaller inserted tube. While for circular tube, $D_H = D$. Therefore, D_H and λ can be scaled accordingly, D_H/λ can be considered as an appropriate dimensionless parameter that defines the detonation limit.

It can be seen from Table 1 that the minimum values of D_H/λ for C_2H_2-2.5O_2-70%Ar and C_2H_2-2.5O_2-85%Ar mixtures in 36-mm inner diameter tube are 0.89 and 0.49. Due to the uncertainties for the cell size estimation at lower initial pressure and the initial pressure increment is 1 kPa, a specific value of D_H/λ means nothing from the aspect of measurement, instead, the average $(D_H/\lambda)_{ave}$ gives a better overall estimation. Therefore, the average value, i.e., $(D_H/\lambda)_{ave}$ for the failure of stable detonation is 0.69. Table 1 also shows the scaling ratio of the hydraulic diameter (D_H) with the cell size (λ) for annular channels. It can be found the limit value of D_H/λ slightly varies with the width of the channel gap (w). In general, the normalized velocity and parameter D_H/λ approximately collapses to a single curve. The above scaling analysis indicates the cell size provides an appropriate length scale for determining detonation limit. Table 1 shows that the $(D_H/\lambda)_{ave}$ at the limit condition for stable mixtures in all the circular tube and annular channels is 0.66, taking into consideration of the measurement accuracy and the slightly uncertainties of the cell size estimation, $(D_H/\lambda)_{ave}$ fits well in both tested mixtures and different geometries.

As stable detonations in highly argon-diluted mixtures can be closely described by the classical ZND model, it is of interest to repeat the analysis based on the characteristic length scale of detonation structure. The ZND induction length is obtained from chemical kinetics computation using CHEMKIN [14] and the GRI MECH3.0 chemical kinetics mechanism in all the present analysis. It was suggested GRI MECH3.0 is an optimized detailed reaction mechanism designed to model small hydrocarbon fuel, especially C_1-C_2. More importantly, it contains argon. This mechanism has been recently validated by Dias et al. [15] through investigating the combustion properties of fuel-oxygen-argon mixtures, and therefore it is suitable for this study. The ZND induction zone length, which can be readily determined from kinetics data in contrast to detonation cell size, represents another appropriate length scale that characterizes the structure of the stable detonation front propagating in heavily argon diluted mixtures. The scaling factors for two stable mixtures are tabulated in Table 1 as well, it can be seen $(D_H/\Delta_I)_{ave}$ at the detonation limits equals to approximately 15 for stable mixtures. It is noteworthy that the ratio between detonation cell size to induction zone length varies two order of magnitude within the detonability limits, therefore the dimensionless ratio of D_H/Δ_I is less universal than D_H/λ, however, $D_H/\Delta_I \sim 15$ can be considered as a roughly estimation for stable mixtures at the limits in the cases that detonation cell sizes are not available.

4 Concluding remarks

The velocity deficit at the near-limits propagation of acetylene-oxygen mixture with high argon dilution in circular tube and annular channels were analyzed in this study. The present experimental results indicate that the average maximum velocity deficit is approximately 14 % of CJ velocity for the highly argon (70% and 80% vol.) diluted in stoichiometric acetylene-oxygen mixtures in the 36-mm inner diameter circular tube and the annular channel gap with different scales (w = 2, 4.5 and 7 mm). Taking into consideration all the uncertainties in determining the cell sizes and identifying the critical limiting pressure, comparison with both experimental results in circular tube and small annular channels suggests the scaling between hydraulic diameter and the cell size (D_H/λ) or ZND induction zone length (D_H/Δ_I) at the detonation limits, average values of 0.66 and 15, respectively.

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