**Shock Waves manuscript No.**
(will be inserted by the editor)

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# Propagation of Near-Limit Gaseous Detonations in Small Diameter Tubes

Submitted: 21 October 2009

**Abstract** In this study, detonation limits in very small diameter tubes are investigated to further the understanding of the near-limit detonation phenomenon. Three small diameter circular tubes of 1.8 mm, 6.3 mm, and 9.5 mm inner diameters, of 3m length, were used to permit the near-limit detonations to be observed over long distances of 300 to 1500 tube diameters. Mixtures with high argon dilution (stable) and without dilution (unstable) are used for the experiments. For stable mixtures highly diluted with argon for which instabilities are not important and where failure is due to losses only, the limit obtained experimentally is in good agreement in comparison to that computed by the quasi-steady ZND theory with flow divergence or curvature term modeling the boundary layer effects. For unstable detonations suppression of the instabilities of the cellular detonation due to boundary conditions is responsible for the failure of the detonation wave. Different near-limit propagation regimes are also observed, including the spinning and galloping mode. Based on the present experimental results, an attempt is made to study an operational criterion for the propagation limits of stable and unstable detonations.

**Keywords** Gas phase detonation · limits · small tubes

**PACS** 47.40.Rs

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## 1 Introduction

Detonability limits refer to the conditions, outside of which the detonation wave fails to propagate [1]. Limits are consequence of the influence of initial and boundary conditions on the propagation mechanism of the detonation wave. The onset of detonation limits depends on a combination of many factors, in general, e.g. tube diameter, the initial thermodynamic state of the mixture $(p, T)$, and the mixture composition, etc., all of which can render the detonation to fail [1].

For small tubes, boundaries introduce heat and momentum losses and these losses can cause detonation velocity deficits and eventually failure. The boundary layer behind a propagating detonation wave leads to flow divergence in the reaction zone, and the effect is similar to that of wave curvature that results in velocity deficits and failure again below a critical curvature of the front. For most gaseous combustible mixtures, detonations are unstable and the instabilities have been known to provide an essential mechanism for detonation propagation. The interference of the tube wall with the intrinsic instability of the cellular detonation front again can lead to limit, for example, when the boundary conditions do not permit even the lowest unstable mode to occur, the detonation fails. Because of all these possible combined effects, it remains difficult to predict the limit and isolate which mechanism dominates to cause failure.

Near the limits, the propagation phenomenon is generally unsteady and very complex. Extensive studies of the limit problem have been carried out in the past. Manson and Guénoche [2] studied explosive mixtures of acetylene-oxygen and propane-oxygen at different compositions, and found that for a given composition, the detonation velocity decreases linearly with the tube diameter as the limit is approached. They proposed that the velocity deficit and limit are results of a quenched layer of mixture in the vicinity of the tube wall due to heat losses. Gordon et al. [3] studied experimentally the phenomenon near the detonation limits in hydrogen-oxygen mixtures in a 20 mm diameter tube for a range of initial pressures and mixture compositions. Their studies indicated that the appearance of the lowest unstable mode of spinning detonation is an indication that detonation limit is approached. Further progress toward the limits after the onset of the spinning detonation regime results in failure. It may be concluded from Gordon's study that instability is an important mechanism in the propagation of a detonation wave and when boundary conditions are such that even the lowest unstable mode is suppressed, the detonation failed. Pusch and Wagner [4] investigated detonation limits in methane-oxygen mixtures and confirmed that the detonability limit is a function of tube diameter, as well as the initial thermodynamic state $(p, T)$, and mixture composition. Their experiments showed the limiting tube diameter is a minimum near the most detonable stoichiometric mixtures. Thus their study implicitly implies that the limiting tube diameter is linked to the reaction zone thickness. Failure occurs when the reaction zone thickness becomes the same order of the tube diameter. The importance of Wagner's work is that it showed detonation limit is not a fundamental property of the explosive mixture only, but also of the boundary conditions.

Manson et al. [5] were perhaps the firsts to point out explicitly that detonability limit is closely related to the stability of the detonation wave. They attempted to define limits on the basis of the local velocity fluctuations from the mean value, which they used as a measure of instability. They studied propane-oxygen-nitrogen mixture and took streak Schlieren photographs of the detonation, which showed increasing scale of the instability of the structure as the limit is approached. They also discovered the galloping detonation mode in certain cases, a periodic failure and re-initiation of the detonation that occurs cyclically over distance of propagation of a hundred tube diameters of travel when limits occur. Galloping detonations were considered as the lowest unstable self-sustained mode of propagation even though during the galloping cycle, the detonation actually failed with the reaction zone decoupled from the precursor shock followed by re-initiation of a detonation. However, galloping detonations are still self-sustained and can propagate for many cycles. Later, St-Cloud et al. [6] investigated in more detail the structure of galloping detonations. They investigated propane-oxygen-60% nitrogen mixture and showed that the reaction zone completely decoupled from the leading shock in the low velocity phase of a galloping cycle prior to re-establishment of the detonation to begin the next cycle of galloping. Thus at the limit the detonation can fail completely and becomes a deflagration and then re-initiate itself again in a galloping cycle.

Because of the effect of boundary conditions, the propagation limits should be governed by the tube dimension and geometry. With the importance of instability in the near-limit conditions, the tube diameter should be correlated to the characteristic length scale of the detonation itself. In Moen et al.'s [7] investigation they proposed that the onset of the spinning mode can be used as the criterion for defining the detonation limit. From the acoustic theory of spinning detonations, Moen et al. arrived at the limit criterion of $\lambda = 2 \cdot d$, where $\lambda$ is the cell size of the detonation. Thus, Moen et al. directly linked instabilities to the detonation limit by proposing a criterion based on the lowest unstable spinning mode. Dupré et al. [8-10] investigated further on the limit criterion and carried on experiments in five detonation tubes of decreasing diameter (152, 97, 74, 49, and 38 mm) at atmospheric initial pressure in lean hydrogen-air mixtures. Their results indicated that most of the near-limit detonations occurred in a region described by $\lambda = \pi \cdot d$, a criterion first proposed by Lee [11]. Lee argued that since $\pi \cdot d$ represents the largest characteristic length scale of the tube it should correlate with the length scale that characterizes the sensitivity of the mixture i.e. detonation cell size $\lambda$. Dupré et al. also showed that the maximum velocity deficit for the last stable mode of propagation corresponded to about 10% velocity deficit from the CJ velocity. Dupré's results indicate that both velocity deficit and instability characterize the detonation limits. Manzhalei [12] studied the limits of galloping detonations in capillary tubes in an acetylene-oxygen mixture and demonstrated the existence of boundary layer stabilized steady waves at velocities as low as half the normal CJ value. Thus the low velocity phase of the galloping cycle can be ascribed to the effect of boundary layer.

Lee et al. [13] developed a Doppler microwave interferometer technique to observe the continuous velocity fluctuations of near-limit detonations in a variety of mixtures. They classified the near-limit propagation phenomenon to consist of a spectrum of unstable phenomena from stable spinning, rapid fluctuation, stuttering, and galloping modes with increasing fluctuation in the velocity. They measured continuously the velocities over a long distance of propagation, of the order of 300 tube diameters.

Recently, Kitano et al. [14] studied the deficits of detonation velocities in hydrogen-oxygen mixtures over long distances of propagation ranging from 300 to 1000 tube diameter distances, and focused on the spinning detonation mode and its transition from multi-head detonations. Chao et al. [15] also investigated the velocity deficits of the near-limit detonations in two-dimensional annular channels for stable argon diluted mixtures. Their study indicates that the maximum velocity deficit at the limits corresponded to 20% from the CJ velocity for hydrogen-oxygen and acetylene-oxygen mixtures diluted with over 50% argon. A more recent work was also performed by Jackson et al. [16] who uses high-speed photography to reveal different regimes of near-limit propagation in very small diameter tubes.

Theoretically, Zel'dovich [17,18] was perhaps the first to attempt to develop a theory for detonation limits based on heat and momentum losses to the tube wall. The detonability limit is reached when no steady solution to the conservation equations described the steady ZND detonation structure is possible at some critical value of the losses. However his model failed to provide a quantitative prediction of the detonation limits. Momentum losses modelled by a body force term in the momentum equation in Zel'dovich's model is not correct even qualitatively. According to Fay [19] the correct interpretation of the heat and momentum losses is due to the presence of a boundary layer. The displacement effect leads to a divergence of the streamlines and hence wave curvature. Curvature can result in a velocity deficit and eventually failure of the detonation when the curvature is excessive and exceeds a certain critical value . Dove and Tribbeck [20] extended Fay's model using more detailed chemical kinetics for the reactions in hydrogen-oxygen mixtures. Some attempts have also been made to correlate detonability limits with the cellular instability of the front [21-22]. However, to-date, there is still no quantitative theory that can predict detonation limits. A result perhaps due to the role of velocity deficit and instabilities not clearly distinguished.

Despite of all these studies, the phenomenon is still not fully understood at present. The important problem of limits has not been clearly resolved [1]. The purpose of this research work is to further extend the investigation of the detonation limit phenomena for a wider spectrum of stable and unstable mixtures over long distances of propagation ranging from 300 to 1500 tube diameter long. Previous studies on detonation limits in tubes were mostly limited to observations over short distances of propagation (less than 300 tube diameters). Only the works of Lee et al. [13], Kitano et al. [14] and Jackson et al.[16] looked at the detonation limit phenomena for distances much more than 300 tube diameter long and thus they were able to observe four distinct near-limit modes of propagation.

In this study, three small diameter circular tubes were used to permit the near limit detonations to be observed over long distances of at least 300 tube diameters. Two types of mixtures were also considered, which was shown in previous studies [23-24] that the detonation is stable ($C_2H_2$ + 2.5 $O_2$ +70% Ar), or unstable ($C_2H_2$ + 2.5 $O_2$ and $CH_4$ + 2 $O_2$). By stable detonations we referred to the propagation mechanism being that of shock induced ignition as described by the classical ZND model. For stable mixture highly diluted with argon for which instabilities are not important and where failure is due to losses only, the limit obtained experimentally can be compared with ZND theories with flow divergence or curvature term to model the boundary layer effects using Fay's model. Unstable detonations referred to those that instabilities and turbulence play the dominant role in the reaction zone. In addition to losses, the existence of detonation limits in the unstable mixture should also be a direct consequence of the interference of the boundary conditions with the inherent instability of the detonation front. Hence, the present study of the limit phenomenon in these two types of mixtures contrast the differences in the propagation and failure mechanisms of stable and unstable detonations. This will perhaps permit a criterion to be defined for detonation limits of stable and unstable detonations.

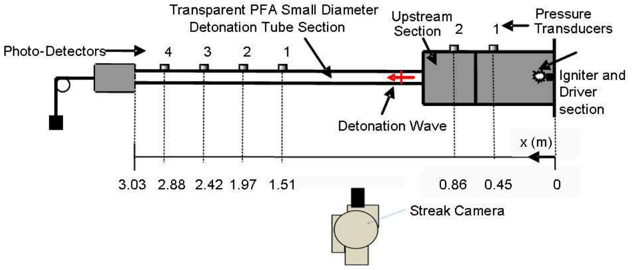

**Fig. 1** A schematic of the experimental setup

## 2 Experimental details

A 1.0 m long steel detonation tube section with an inner diameter of 63.5 mm was used to ensure that a steady CJ detonation is initiated prior to its entering in the smaller diameter Perfluoroalkoxy (PFA) transparent test tube of interest. The near limit detonation phenomenon was observed over a 3.0 m long distance. One end of the (PFA) transparent tubing was affixed to the end of the upstream initiator steel section and the other end terminates in a steel cylinder where hanging weights are used to apply tension to the thin plastic tube to keep it straight. Tubing of different diameters were considered ($d$ = 1.8 mm, 6.3 mm and 9.5 mm). With these small diameter tubings, the detonation can be observed over a propagation distance of $L/d$ = 1666, 476, and 315 respectively. A sketch of the experimental apparatus is shown in Figure 1. A gas control panel, equipped with an Omega pressure transducer (PX02-I) and a Newport digital meter (IDP) was used to monitor the initial pressure in the detonation tube for the experiment. Mixtures of $C_2H_2$ + 2.5 $O_2$ +70% Ar, $C_2H_2$ + 2.5 $O_2$ and $CH_4$ + 2 $O_2$ were first premixed in storage vessels by the method of partial pressure and allowed to mix by diffusion over 24 hours before use. When the mixtures were ready to be used, the detonation tube was evacuated to at least 80 Pa. Then, in order to ensure an uniform gas distribution in the PFA tubing section, the detonation tube was overfilled from upstream, and evacuated from downstream to the desired test pressure. In all of the present experiments, the sensitivity of the explosive mixture was varied to bring about the onset of limits by changing the initial pressure, $P_o$, while the mixture composition was kept fixed. Thus, the initial pressure is the principal parameter that varies to determine the detonation limit for each of the three mixtures and tube diameters.

A powerful spark from a capacitor discharge was used to ignite the mixture and generate the detonation by direct initiation in the steel tube section. The detonation was then transmitted to the smaller test tube. For cases of very insensitive mixtures near the limit, where direct initiation by the energy spark cannot be achieved, a driver section filled with stoichiometric acetylene-oxygen at 30 kPa, separated initially from the upstream steel section by a thin mylar diaphragm, was used to initiate the insensitive test mixture.

Two piezoelectric pressure transducers (PCB 113A24) were mounted on the driver and the steel detonation tube section in order to verify that a CJ detonation was obtained prior to its transmission to the smaller PFA tubing. Also, for measuring the detonation velocity in the plastic tubing, ten 1 mm diameter optic fibers connected to a photodiode (IF-95OC) were spaced periodically along the PFA transparent tubing to determine the time of arrival, and hence the detonation speed. From the time of arrival of the detonation at the different locations along the PFA tubing, the speed of the detonation can be calculated. A rotating drum streak camera with a film speed of about 83 m/s was also used to provide a continuous observation of the propagation of the detonation wave in the PFA test tube. The speed of the detonation wave can be calculated from the slope of the luminous streak.

## 3 Experimental results

Experiments using the three different mixtures (acetylene-oxygen mixture, methane-oxygen mixture, and acetylene-oxygen-70 % argon mixture), and the three different tube diameters (9.5 mm, 6.3 mm, and 1.8 mm) were carried out to investigate the propagation of near-limit detonations. The initial pressure was decreased progressively to bring about the onset of detonation limit and eventually failure of the detonation for each mixture at a given tube diameter. For the various mixtures and tube diameters, the propagation of the detonation shares similar features as the detonation limits are approached. Therefore, the experimental results will be presented to highlight their common characteristics and differences.

### 3.1 Conditions well within the limits

Figure 2 shows a typical streak photograph for the case where the conditions are well within the detonation limits (i.e., when the initial pressure is far above the critical limiting pressure). In general, the detonation is found to be slightly overdriven upon exiting the steel section into the smaller diameter test section. This is due to the high pressure from the reflection of the detonation upstream in the larger diameter steel tube which overdrives the detonation in the smaller tube initially. However, the overdriven detonation decays rapidly and propagates subsequently at a constant velocity for the remaining length of the tube. It can be observed also that the detonation velocity measured from the signal of photodetectors and the slope of the streak is very close to the theoretical CJ value with velocity deficits typically of the order of a few percent (about 3-5%). Similar results are obtained for all three tube diameters and mixtures considered.

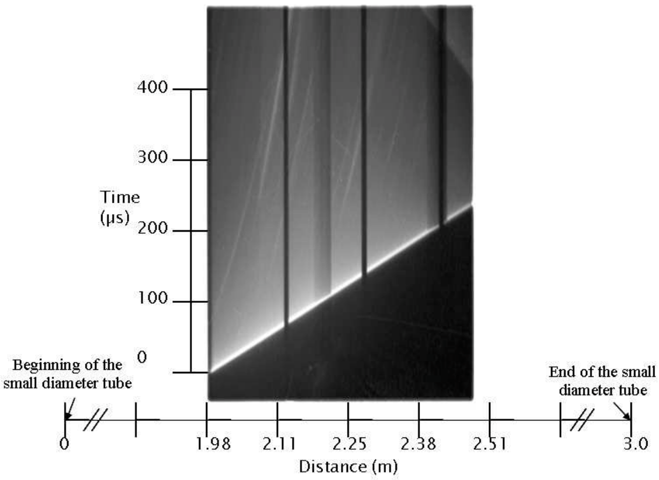

**Fig. 2** $C_2H_2/O_2$ mixture in the 9.5 mm diameter tube at 10 kPa

### 3.2 Conditions outside the limits

For the other extreme cases when the initial pressure is significantly less than the critical value, the detonation wave is found to decay progressively from its initial overdriven state to a turbulent deflagration wave as it propagates down the tube, as shown from the streak photographs in figures 3 and 4. We referred the condition resulted in the detonation decay to a deflagration to be outside the detonation limits. The eventual turbulent deflagration velocity is found to be typically about 60% the theoretical CJ detonation velocity near the end of the 3 m tube. The decay appears to be slow and not abrupt since the 3 m length of the tube corresponds to about 300 diameters even for the largest diameter used of 9 mm. The high speed deflagration observed when the detonation fails appears to correspond to the metastable deflagration speed of about half the CJ detonation velocity, during the transition from deflagration to detonation reported by Zhu et al. [25].

For the undiluted mixture ($C_2H_2$ + 2.5 $O_2$) in the 1.8 mm diameter tube, it may be of interest to point out one particular phenomenon for the case of smallest diameter tube wall as shown in figure 5. The detonation decays and then manages to continue to propagate at an average velocity of about 83% of the theoretical CJ velocity value for almost the entire length of the 3 m long tube towards the end, (i.e a distance of about 1000 tube

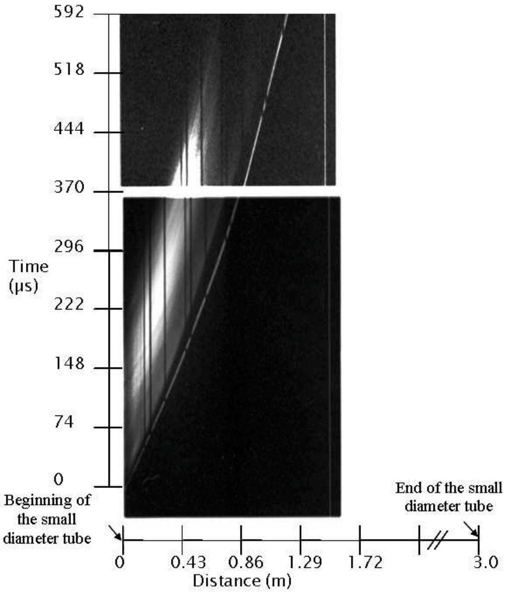

**Fig. 3** $C_2H_2/O_2$ mixture in the 9.5 mm diameter tube at 0.8 kPa

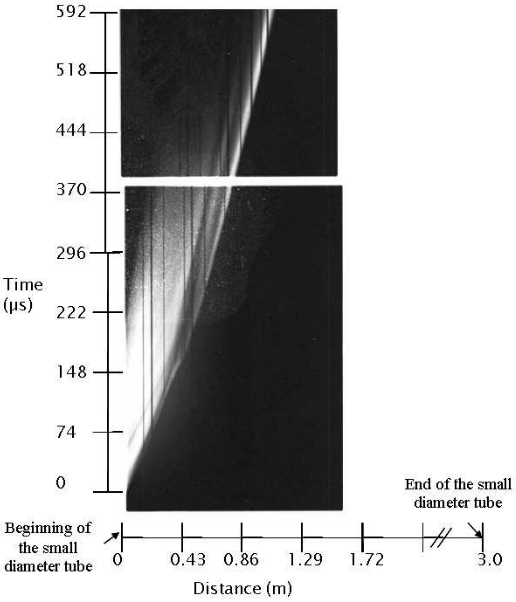

**Fig. 4** $CH_4/O_2$ mixture in the 6.3 mm diameter tube at 5 kPa

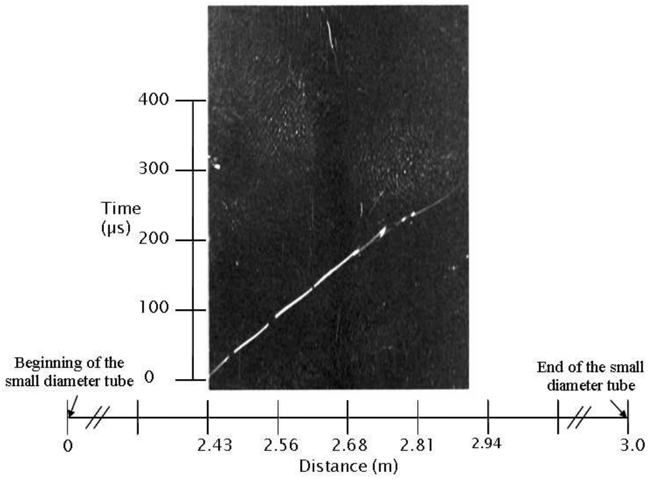

**Fig. 5** Streak photograph of a failing detonation wave with $C_2H_2/O_2$ mixture in the 1.8 mm diameter tube at the critical pressure of 5 kPa

diameters). We also note that the velocity has large local fluctuations but the averaged speed is closer to the CJ detonation speed than to the deflagration speed. This regime can be called the quasi-detonation regime and since it is only observed in the smallest diameter (i.e. 1.8 mm) tube, the boundary layer is probably playing an influential role in stabilizing the wave. A theoretical model for this boundary layer stabilized quasi-detonation was given by Manzhalei [12] who studied detonation propagation in capillary tubes. Thus, boundary layer stabilized detonations should not be considered as normal detonations and the limit criterion for larger diameter tube may not be relevant to the 1.8 mm tube.

### 3.3 Conditions near the onset of the limits

As the detonation limit is approached by decreasing the initial pressure to some critical value, both the structure and the detonation velocity change rapidly. We shall refer to this condition as the near-limit condition. The exact value of the limiting pressure is difficult to determine due to the unstable and irreproducible nature of the near limit phenomenon. Numerous experiments would be required to obtain a meaningful statistical basis for analysis. In the present study no attempt was made to iterate for the exact value of the limiting pressure.

From the streak photographs in Figures 6 and 7 we can observe the typical phenomena of the unstable detonation structure at the onset of detonation limits in both the undiluted and the diluted acetylene mixtures, i.e. $C_2H_2$ + 2.5 $O_2$ mixture, and $C_2H_2$ + 2.5 $O_2$ + 70% Ar mixture, for different tube diameters. It can be seen that at the onset of limits, the detonation wave manifests itself as a spinning detonation. The periodic structure is quite evident from the streak photographs. The velocity deficits also increase and for steady spinning detonations these are of the order of 15% of the theoretical CJ velocity value, whereas, normally the velocity deficits are of the order of a few percent in the same tube when the mixture is well within the limits.

For the unstable mixture $CH_4$ + 2 $O_2$, it can be observed that past the spinning mode, the detonation wave can also propagate as a galloping detonation (Figures 8 and 9). The galloping detonation mode is characterized by a periodic failure and re-initiation of the det-

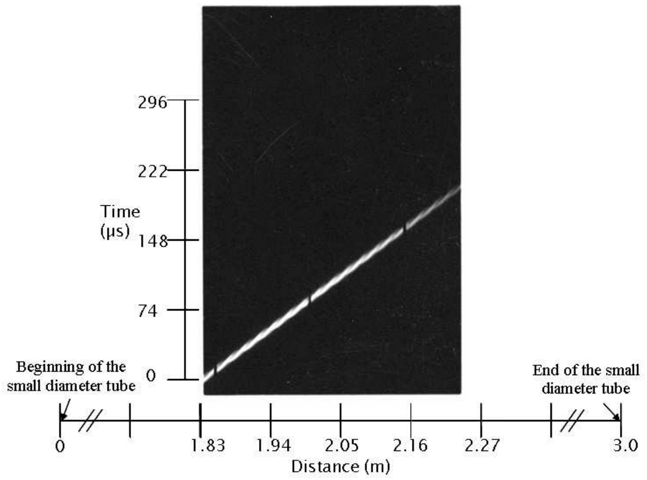

**Fig. 6** Streak photograph of a spinning detonation wave in $C_2H_2/O_2$ mixture in the 6.3 mm diameter tube at the critical pressure of 1.5 kPa

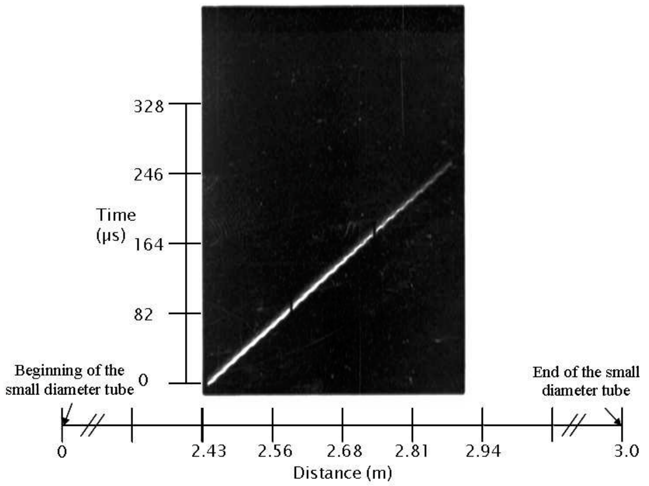

**Fig. 7** Streak photograph of a near-the-limits detonation wave with $C_2H_2/O_2/70\%Ar$ mixture in the 6.3 mm diameter tube at the near-limit pressure of 6 kPa

onation wave. The cyclic fluctuation of the velocity of the galloping detonation wave varies between 40% the theoretical CJ velocity value in the low velocity phase of the galloping cycle to above the theoretical CJ velocity value immediately after re-initiation. In a galloping detonation, the detonation actually failed with the reaction zone completely decoupled from the leading shock as in a deflagration. The velocity of this phase of the galloping cycle is less than half the CJ value. However, the detonation re-initiates itself at the end of the low velocity cycle. It is arguable that galloping detonation can be called "detonation" since a cycle occurs over a distance of many tube diameters. However, the averaged velocity of galloping detonation can be close to the CJ value in spite of the large fluctuations. Furthermore, the phenomenon is self-sustained. In general, we can see that

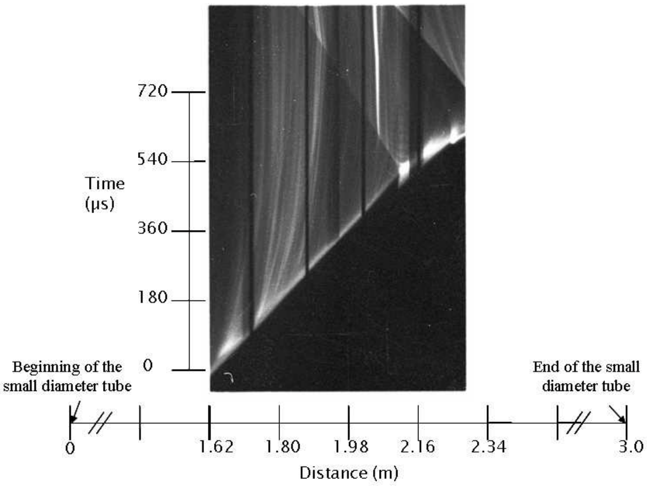

**Fig. 8** Streak photograph of a galloping detonation wave in $CH_4/O_2$ mixture in the 9.5 mm diameter tube at the near-limit pressure of 10 kPa

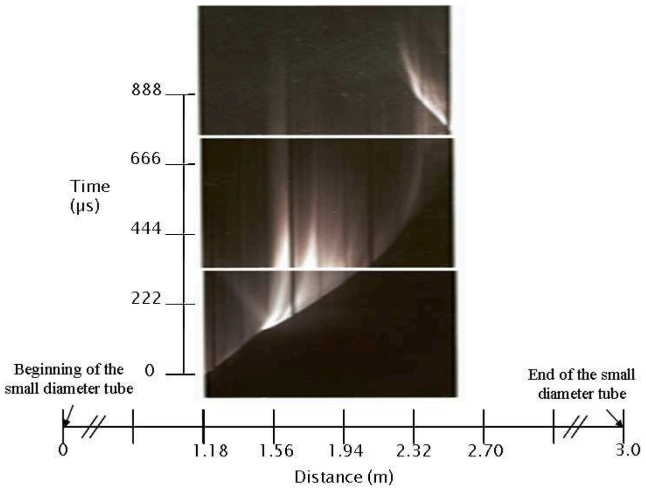

**Fig. 9** Streak photograph of a galloping detonation wave in $CH_4/O_2$ mixture in the 6.3 mm diameter tube at the near-limit pressure of 18 kPa

for near-limit conditions, the velocity deficit increases to about 20% whereas for detonations well within the limits, the velocity deficit is usually of the order of a few percent. The onset of limits corresponds to the appearance of spinning and galloping detonations as has been well documented in previous studies (Gordon et al. 1959, and Dupré et al. 1985 ,etc.). Single headed detonations in round tubes correspond to the lowest unstable mode of the detonation. This led Moen (1985) to propose the use of the onset of single headed spin as a criterion to define the limits.

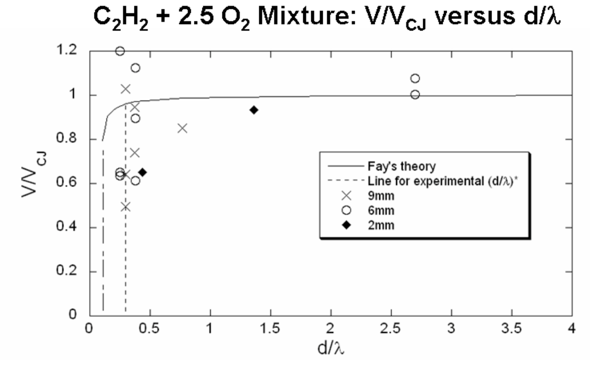

**Fig. 10** Detonation velocity (normalized with its theoretical CJ velocity value) as a function of the detonation limit criterion, $d/\lambda$, for the $C_2H_2$ + 2.5 $O_2$ mixture.

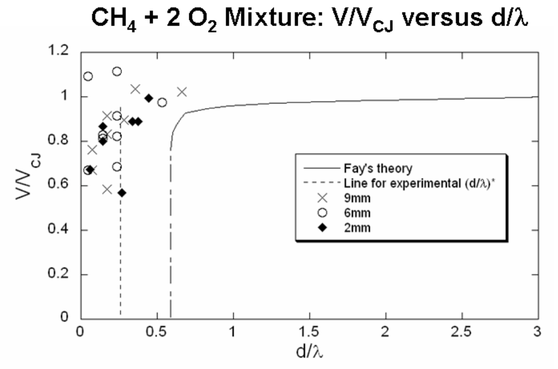

**Fig. 11** Detonation velocity (normalized with its theoretical CJ velocity value) as a function of the detonation limit criterion, $d/\lambda$, for the $CH_4$ + 2 $O_2$ mixture.

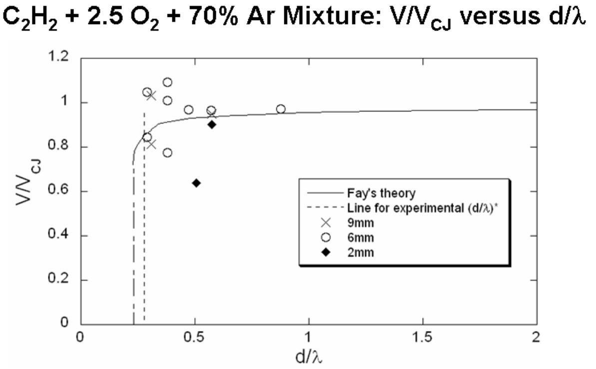

**Fig. 12** Detonation velocity (normalized with its theoretical CJ velocity value) as a function of the detonation limit criterion, $d/\lambda$, for the $C_2H_2$ + 2.5 $O_2$ + 70% Ar mixture

## 4 Analysis and discussion

Experimental data for the cell size of the three mixtures used in the present study, i.e. $C_2H_2$ + 2.5 $O_2$ mixture, $CH_4$ + 2 $O_2$ mixture, and $C_2H_2$ + 2.5 $O_2$ + 70% Ar mixture, as a function of initial pressures has been collected/correlated from the GALCIT detonation database [26]. Whereas the cell size, $\lambda$, is the length scale that characterizes the sensitivity of the explosive mixture, the tube diameter, $d$, is the physical length scale of the boundary condition that determines the detonation limit (for a given mixture at a given initial pressure). Thus, the ratio $d/\lambda$ should be an appropriate dimensionless parameter that defines the detonation limit.

The velocity deficit results obtained from experiments are plotted as a function of $d/\lambda$, in figures 10-12, for all the different mixtures in the three different tube diameters investigated. To compare the experimental results and elucidate the failure mechanism, the velocity deficits for the various mixtures and initial pressures are computed using Fay's model and are also shown in the same plots. The velocity deficits and limits are calculated theoretically by solving the quasi-one-dimensional ZND structure equations with a curvature term and detailed chemical kinetics for the reactions. In the quasi-steady ZND formulation, the wall effect is modelled by the negative displacement thickness of the boundary layer behind the leading shock front following the pioneering work of Fay [19]. The effect of the viscous boundary layer tends to remove mass from the free stream flow and is accounted for by the flow of an inviscid fluid through an expanding nozzle where the cross-sectional area change is related to the displacement thickness of the boundary layer developed behind the shock front. The effective cross-sectional area of the tube at any point is given by $A = \pi((d + 2\delta^*)/2)^2$. The area change and the mass displacement thickness $\delta^*$ are given as [27]:

$$\xi(x) = \frac{1}{A}\frac{dA}{dx} = \frac{4}{d+2\delta^*}\frac{d\delta^*}{dx}$$
$$\delta^* = 0.22 \cdot x^{0.8}\left(\frac{\mu}{\rho_o D}\right)^{0.2}$$

The ZND detonation structure is described by the solutions of the following system of ordinary equations:

$$\frac{dp}{dt} = -\rho u^2 \frac{\dot{\sigma} - u\xi}{\eta}$$
$$\frac{d\rho}{dt} = -\rho \frac{\dot{\sigma} - uM^2\xi}{\eta}$$
$$\frac{du}{dt} = u\frac{\dot{\sigma} - u\xi}{\eta}$$
$$\frac{dy_i}{dt} = \frac{W_i\dot{\omega}_i}{\rho}(i = 1 \ldots N_s)$$
$$\frac{dA}{dt} = Au\xi$$

with

$$\eta = 1 - M^2$$
$$\dot{\sigma} = \sum_{i=1}^{N_s}\left(\frac{W}{W_i} - \frac{h_i}{c_p T}\right)\frac{dy_i}{dt}$$

where $\rho, A, u, p, T, h, N_s, y_i, W_i, \dot{\omega}_i$ denote mixture density, cross-sectional area, particle velocity, pressure, temperature, enthalpy, total number of species, mass fraction, molar mass and molar rate of production of species $i$. $M$ is the Mach number of the flow relative to the shock wave, $W$ is the mean molar mass of the mixture, $c_p$ is the mixture specific heat at constant pressure, and $h_i$ is the specific enthalpy of specie $i$.

These ZND model equations [28] with the flow divergence term in the conservation of mass equation and together with the chosen detailed chemistry model are solved numerically to seek the eigenvalue solution satisfying the generalized CJ criterion in order to achieve a continuous transition through the sonic plane (i.e. when the flow is choked in the frame of reference of the leading shock, the rate of chemical energy release must balance the rate of mass divergence). The system of equations was numerically integrated using the CHEMKIN II package [29]. Following the methodology discussed in [28-31], different detonation velocities were iterated to determine numerically the eigenvalue solution when the flow condition attained $M = 1$ in a continuous regular transition. The limit is achieved when the velocity deficit exceeds a certain maximum value (or a certain critical curvature of the front) and no steady ZND solution can be obtained. For both the acetylene-oxygen-argon and methane-oxygen mixtures, the UC San Diego reaction kinetic mechanism was used [32-33]. From the experimental data, the velocity deficit increases sharply near a critical value of $d/\lambda$, thus defining the limits.

In Figure 10 showing results for $C_2H_2$ + 2.5 $O_2$, the limiting value of $d/\lambda$ from Fay's theory is found to be about 0.1, whereas the experimental data suggest $d/\lambda \approx 0.3$, which is in agreement with the single headed spin criterion. In Figure 11 for $CH_4$ + 2 $O_2$, Fay's theory predicts a value of $d/\lambda = 0.5$, whereas the experimental data indicates $d/\lambda = 0.3$, again in better accord with the criterion $d/\lambda$=1/$\pi$, as in $C_2H_2$ + 2.5 $O_2$ mixtures. In the stable mixtures of $C_2H_2$ + 2.5 $O_2$ + 70% Ar it appears that both theory and experiments are in better agreement with each other (see figure 12). Note that in highly diluted mixtures with argon, the detonation is stable and thus falls under the one-dimensional ZND model description. Thus, one would expect the experimental results in a better agreement with Fay's quasi-1D model. For the cases of unstable mixtures, the experimental results seem to deviate more from the quasi-one-dimensional model of Fay which is essentially based on the steady ZND model. For the detonations in the unstable mixture of methane-oxygen, the same near-limit modes of propagations, as those observed previously by authors such as Lee et al.(1995), and Kitano et al.(2008), are recovered here, in the present research work. Also, for detonations in the unstable mixture of methane-oxygen, it is found that the suppression of the instabilities due to boundary conditions is a more appropriate mechanism responsible for the failure of the detonation wave. It is important to point out that since no attempt were made to iterate the precise value of the critical pressure experimentally, it is not possible to obtain a more accurate value of $d/\lambda$ at the limit for the various cases studied.

## 5 Conclusion

In this work the near limit phenomenon of detonation propagation in round tubes is investigated. Small tube diameters (compared to the tube length) are used to permit the near limit behaviour to be observed over large $L/d$ ratio ($300 \leq L/d \leq 1500$). Also two types of mixtures, which are known to give stable and unstable detonations are used. Thus, the limit behaviour for these two types of detonations can be compared. The propagation phenomenon of the detonation wave was observed with a rotating drum streak camera over many tube diameters to define the failure condition. Also optic fibers periodically spaced along the PFA transparent tubing were used to obtain an accurate measurement of the detonation speed. As the initial pressure of the mixture was varied to bring about the onset of detonation limits, the detonation wave was observed to share distinguished characteristics in different ranges of initial pressure. First, for all three mixtures tested in all three tube diameters, it is found that when the condition is well within the limits (i.e., when the initial pressure is far above the critical limiting pressure), the detonation velocity is found to be constant throughout the whole length of the tube, and the steady detonation velocity is shown to have velocity deficit that depends on the tube diameter. When the initial pressure is near the critical pressure (referred to as the near limit condition), the streak photographs in both the undiluted and the diluted acetylene mixtures (i.e. $C_2H_2$ + 2.5 $O_2$ mixture, and $C_2H_2$ + 2.5 $O_2$ + 70% Ar mixture) show that spinning detonations occur near the onset of detonation limits. For spinning detonations the velocity deficits are still of the order of 15% of the theoretical CJ velocity value. For the unstable detonations in mixture $CH_4$ + 2 $O_2$, it is found that past the spinning mode, the detonation wave continues to propagate as a the galloping detonation. The cyclic velocity fluctuation of the galloping detonation wave can vary between from 40% the theoretical CJ velocity value in the low velocity phase of the galloping cycle to above the theoretical CJ velocity value after re-initiation. When the initial pressure is significantly below the limiting value, the detonation wave is found to decay progressively from its initial overdriven state to a turbulent deflagration wave as it propagates down the tube. The turbulent deflagration velocity is typically about half the theoretical CJ detonation velocity, similar to the metastable deflagration prior to the transition from deflagration to detonation.

The velocity deficits for the various mixtures and initial pressures were computed from Fay's model and compared with the experimental detonation velocities (normalized with respect to the theoretical CJ value). For both the unstable mixtures tested, the limiting value of $d/\lambda$ predicted by Fay's model deviates from the experimental value which appears to agree better with the criterion of $d/\lambda = 1/\pi$. Thus, it appears that the onset of single headed spin criterion where $d/\lambda = 1/\pi$ seems to describe the limiting condition for steady propagation of detonation in these cases. On the other hand, in stable argon diluted mixture, the results from the theory of Fay agrees well with the experiments.

Although the present study presents a series of new experimental results, it may still not be sufficient for an accurate determination of the value of the limiting pressure for the different cases analyzed. A more definitive study would require a larger data base of velocity data so that a meaningful statistical value of the limiting pressure can be precisely identified.

**Acknowledgements** This work was supported by the Natural Sciences and Engineering Research Council of Canada (NSERC).

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