Velocity Fluctuation Near the Detonation Limits

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Abstract

In this study, the velocity fluctuation near the detonation limits is investigated experimentally. Five explosive mixtures in five different diameter tubes were used and the choice of the mixtures included those considered as "stable" with regular cellular pattern and "unstable" with highly irregular cellular pattern. Photodiodes spaced at regular intervals along the tube were used to measure the detonation velocity. Piezoelectric transducers were also used to record the pressure profiles. Smoked foils were used to register the cellular detonation structure. Away from the limits, the detonation is found to propagate at a steady velocity throughout the length of the tube and the fluctuations of the local velocity are generally small. For stable mixtures with high argon dilution, the onset of the detonation limits is indicated by an abrupt drop in the detonation velocity to about $0.4V_{CJ}$ after a short distance of travel. The detonation may continue to propagate at this low velocity before decaying eventually to a deflagration wave. For deflagrations the optical detector sometimes failed to register a signal due to low luminosity of the front. In unstable mixtures, galloping detonations are observed only in small diameter tubes (e.g., D = 12.7, 3.2 and 1.5 mm). A large number of fairly reproducible cycles of galloping detonations can be observed in very small diameter tubes. In large diameter tubes (e.g., D = 31.7 and 50.8 mm), no galloping detonations are observed in all stable and unstable mixtures. For stable mixtures, no galloping detonations are observed even in small diameter tubes of D = 3.2 and 1.5mm. Smoked foils records show that the cellular detonation structure changes from multi-headed to single-headed spin as the limit is approached. In a galloping detonation cycle, a decay from multi-headed to single-headed detonation is observed. However, the cellular structure vanishes for further decay of the galloping detonation to the low velocity phase of the galloping cycle. Although galloping detonations could be considered to define the boundary for detonation limits, this definition lacks generality since galloping detonations are not always observed in all mixtures and in all tube diameters. Thus the onset of single-headed spin is perhaps the most appropriate criterion of the detonation limits in tubes.

Keyword: Detonation limits; Velocity fluctuation; Galloping detonation; Single-headed spinning detonation; Cellular structure

1 Introduction

Detonation waves are intrinsically unstable with a transient cellular structure formed by an ensemble of interacting transverse waves. Thus the local velocity of the detonation front fluctuates about a mean value of the order of the CJ velocity with a frequency inversely proportional to the cell size. Further decrease in mixture sensitivity leads to the enlargement of the cell size (or transverse wave spacing) and the detonation limit is approached, i.e., conditions outside of which the detonation wave fails to propagate.

The detonation velocity near the limits is reported in a previous paper [1]. The velocity is a value averaged over the distance of propagation of the detonation wave. However near failure of the detonation, the velocity fluctuations become increasingly large rendering the averaged velocity of doubtful significance. In fact, the failure mechanism is obscured in the averaging process since it is the instability itself that is responsible for the propagation of the detonation wave. Therefore, to understand detonation limits, one must investigate the instability of the front as the limits are approached. The present study emphasizes the instability of the front as the limits are approached.

It is well known that far from the limits the frequency of the transverse instability is high (or equivalently the cell size is small); the instability tends toward lower modes and eventually single-headed spinning detonation is reached. While the scale of the frontal instability is of the same order as the tube diameter ($\lambda \approx \pi \cdot D$), in fact the onset of single-headed spinning detonation had been chosen to define the detonation limits by various investigator, e.g., [2-4]. The single-headed spinning detonation represents the lowest mode of transverse instability of the detonation front. However, numerous investigators have reported

longitudinal instability in the form of "stuttering" and galloping detonations; see [4-9] and references therein. The velocity fluctuation of these longitudinal instabilities ranges from as low as $0.4V_{CJ}$ to $1.2V_{CJ}$ and the detonation of the low velocity phase (i.e., $0.4V_{CJ}$) can be very long. A galloping cycle is generally over 300 tube diameters, yet the averaged value of the velocity for galloping detonations is found to be still close to the CJ value. Thus, it appears that galloping detonation waves have similar apparent characteristics of a genuine detonation and that the limits should be extended beyond the spinning mode to include these longitudinal unstable detonations. In fact, a number of investigators have suggested that galloping detonation be considered as boundary for the onset of detonation limits. Beyond galloping detonations, the detonation velocity decayed to about $0.4V_{CJ}$ and this is generally referred to as "low-velocity detonation" or sometimes it is also called high-speed deflagration. Whether these quasi-steady, low-velocity detonations or fast deflagrations can be considered as detonations is not clear. In addition, little is known about their structures. The mechanisms of galloping and low-velocity detonations are not understood and relatively little detailed study of these unstable detonations waves with large fluctuations had been made. It appears that this class of longitudinal unstable detonations is crucial towards understanding of the failure mechanisms at the detonation limits since they occur just prior to failure.

In our continuing effort to understand the detonation limits phenomenon, the present study focuses particularly on the velocity fluctuations just prior to failure. Not all detonations in different mixtures and tube diameters exhibit large fluctuations near the limits. Hence in the present investigation, a variety of mixtures, from the high argon-diluted "stable" mixtures to the "unstable" mixture of methane-oxygen, propane-oxygen and fuel-N_2O as oxidizer are

studied. The detonation stability of a mixture is typically assessed by its chemical activation energy or more recently by the stability parameter χ - defined by the activation energy of the induction zone multiplied by the ratio of induction zone to the exothermic heat release length, see [10, 11]. An unstable mixture (e.g., undiluted hydrocarbon fuel mixture) usually has a very large value of activation energy (or stability parameter χ) and the detonation cellular structure in the unstable mixture is observed to be irregular. For a stable mixture, the chemical reaction is less sensitive to any temperature fluctuation. In other words, it has a low activation energy and also a larger heat release region that results in a low value of the stability parameter χ. The detonation structure in stable mixtures is piecewise laminar without any sub-scale instability at the detonation front. Such mixture can be formed with large amount of argon dilution [12].

Since the cycle of the unstable oscillations can cover a large distance of propagation (e.g., hundreds of tube diameter), small diameter tubes as low as 1.5 mm diameter have to be used to ensure propagation over hundreds of tube diameters. Velocity measurement is the main diagnostic used and where possible smoked foils were used to record the cellular detonation structure.

2 Experimental setup

The detonation tube used in the present study consists of a 1.3 m long steel driver section with a diameter of 65 mm. The transparent polycarbonate test tubes of various diameters were attached to the end of the driver tube. Five different diameters, D = 1.5, 3.2, 12.7. 31.7 and 50.8 mm, were used in the present study with total tube length L = 2438, 2438, 4118, 4118, 4118 mm and thickness T = 0.8, 1.6, 3.2, 3.2, 3.2 mm, respectively. The total length L

of the test section was obtained by connecting several same diameter tubes of about 200 mm long using Swagelok tube fittings. Care was exercised to connect smoothly the tubes to avoid any influence of the joint. Detonation was initiated by a high energy spark discharge. A short length of Shchelkin spiral was also inserted downstream of the spark plug to promote detonation formation. For experiments with the small diameter tubes of 1.5 mm and 3.2 mm in diameter, a driver section of 25.4 mm diameter and 1.5 m long with a much more sensitive mixture was used to facilitate the detonation formation and its initial propagation in the test gas before the boundary effect started to take place. A schematic of the experimental apparatus is shown in Fig. 1a.

Five explosives mixtures, i.e., $C_2H_2 + 2.5O_2 + 85\%Ar$, $C_2H_2 + 2.5O_2 + 70\%Ar$, $C_2H_2 + 5N_2O$, $C_3H_8 + 5O_2$, $CH_4 + 2O_2$ were used and the choice include those mixtures considered as “stable” with regular cellular pattern and “unstable” with highly irregular cell pattern. In general, stoichiometric mixtures of acetylene-oxygen with high argon dilution of 85% and 75% argon dilution are considered as "stable" mixture whereas the other three mixtures are considered as "unstable" with irregular cell pattern. The explosive mixtures of the desired composition were prepared via partial pressure in separate gas bottles. The gases were allowed to mix in the vessel by diffusion for at least 24 hr in order to ensure homogeneity prior to being used. For any given experiment, the detonation tube was evacuated to at least 10 Pa. The entire apparatus was then filled from both ends to the desired initial pressure. A gas control panel, equipped with an Omega pressure transducer (PX02-I) and a Newport digital meter (IDP) was used to monitor the pressure for both mixture preparation and the experiment.

Two piezoelectric pressure transducers (PCB 113A24) were mounted on the steel driver section in order to verify that a CJ detonation was obtained prior to its transmission to the test section tube. Fiber optics of 2 mm in diameter connected to a photodiode (IF-95OC) were spaced periodically along the entire length of the test section. Local detonation velocity was measured from the time-of-arrival of the detonation at two neighboring optical probe locations. Typical output from the optical detectors is shown in Fig. 1b. Smoked foils were also used to observe the structure of the detonation in the larger diameter tubes. The smoked foil was made of a thin (300μm) plastic sheet covered with uniform soot and carefully inserted into the test tube before each shot.

3 Results and discussions

All velocity measurements are summarized in Figs. 2 to 7. To ensure the reliability of the results, experiments at the same initial condition were repeated several times. In most cases shown in these figures, different shots at the same initial conditions are also included on the same plot using different symbols to demonstrate their good repeatability.

In general, for a given mixture and a given tube diameter, the detonation limits are approached by progressive decrease in the initial pressure. Above the limits, the detonation velocity remains fairly constant throughout the distance of propagation. Although the local velocity shows some fluctuations as the limits are approached, these are sufficiently small and a meaningful averaged velocity can be obtained. Away from the limits, the average velocity is generally found to be in the range of $0.8\ V_{CJ} \leq V_{avg} \leq V_{CJ}$. The velocity depends slightly on the tube diameter and the mixture. For instance, the velocity deficit increases with

decreasing tube diameter. This qualitative behavior of the velocity deficit is in accord with other previous studies, e.g., [1, 13], and can be described by the Fay-Dabora model [14] as shown in [9, 15].

For the stable mixtures (e.g., $C_2H_2 + 2.5O_2$ with 70% and 85% argon dilution), of which the cellular pattern is rather regular, the onset of limits is indicated by an abrupt drop in the detonation velocity (after a short distance of travel) to a "low-velocity" detonation with a velocity of about $0.4V_{CJ}$. The detonation may show some slight fluctuations in some cases. The slight acceleration of the low-velocity detonation (i.e., $V \approx 0.4V_{CJ}$) is usually observed in very small diameter tube (e.g., 3.2 mm), as shown in Figs. 2c and 2d. In order to verify that there is no other propagation mode such as galloping detonation in stable mixtures, experiments were repeated at P_o = 9kPa using a test section extended about twice the length and the result is given in Fig. 3. In the extended long tube, similar wave acceleration is observed. However, after the acceleration, detonation fails completely and for the low velocity deflagration, the optical probes fail to register due to insufficient luminosity. No cyclic galloping detonation is observed. The observed acceleration could perhaps be due to the wall boundary effects, where the leading shock-boundary layer interaction may cause local instability and ignition resulting in the apparent wave acceleration. However, as shown in Fig. 3, such mechanism is insufficient to sustain any galloping mode of the detonation in stable mixtures. In larger diameter tubes (i.e., 31.7 or 50.8 mm), no acceleration is observed (see Fig. 2).

For an unstable mixture like $C_3H_8 + 5O_2$, the behavior of the detonation near failure is more interesting. Figure 4a shows the velocity for the largest tube diameter D = 50.8 mm.

Above the limiting pressure, the detonation velocity is fairly constant along the length of the tube although some small fluctuations can be seen. The averaged velocity is of the order of $V_{avg} \approx 0.9V_{CJ}$. Approaching the limits, the velocity decreases continuously. Similar to the stable argon-diluted mixtures, galloping detonations are not observed in the larger diameter tubes of D = 31.7 and 50.8 mm). Past the limiting condition, the velocity decays continuously (see Fig. 4b). For the smaller diameter tubes, the near-limit phenomenon becomes more interesting. Large velocity fluctuations are observed (e.g., galloping detonation) and illustrated in Fig. 4c-4f. Many cycles of galloping detonation can be observed in the smallest 1.5 mm diameter tube, whereas in larger diameter tubes, only one cycle of galloping detonation is observed in $C_3H_8 + 5O_2$. It appears that small diameter tubes promote and maintain cyclic fluctuations of the detonation more readily. Note that the cyclic behavior of galloping detonation is fairly reproducible.

By examining the trend of the present results and it seems to indicate that re-acceleration to the overdriven phase of the galloping detonation is not likely in the cases of larger tube diameters. Nevertheless, a remark should be made that the galloping detonation could perhaps exist within a very narrow range of initial pressure and is just not being captured in the present experiment. In addition, the observed galloping cycles typically have periods of about 300 tube diameters. Hence, in the present study another possible reason for the absence of galloping detonations in larger diameter tubes may be due to the insufficient tube length to observe more than one or two cycles. Hence, there remains a possibility that if a longer tube is available, galloping detonations can exist in fairly large tube diameters. However, this remains unclear and future work is necessary to investigate this phenomenon.

The stability of galloping detonations indicates that they are longitudinal instabilities analogous to the pulsating detonations observed in numerical simulations, see [4, 16]. If the galloping cycle is a periodic decay of an overdriven detonation to a decoupled shock and flame in the low velocity phase of the cycle followed by a DDT process from deflagration to detonation, then the galloping phenomenon would be more random (i.e., irregular cyclic period or fluctuation) since DDT is generally a highly irreproducible process. More specifically, the transition or DDT run up distance is highly irreproducible due to the array of turbulent and instability mechanisms that play the role in effecting transition to detonation. Experiments and simulations have led to a general acknowledgment that DDT is effected from rapid turbulent mixing enhanced by shock-flame interaction, flame-vortex interaction and frontal instabilities, etc. These effects are responsible for flame acceleration and formation of "hot spot" or local explosion triggering the onset of detonation. However, the reproducible cyclic oscillation observed here suggests that galloping detonation is more of a gas dynamic phenomenon of non-linear coupling between chemical reactions and the gas dynamic flow field. The absence of galloping detonations in high argon-diluted stable mixtures supports this view.

For the unstable mixture like $CH_4 + 2O_2$, similar behavior as in $C_3H_8 + 5O_2$ is observed (see Fig. 5). Again, in the smallest diameter tube of 1.5 mm, many cycles of galloping detonations are observed. For the unstable $CH_4 + 2O_2$ mixture, the galloping mode is observed over a large pressure range. Note that in the larger diameter of D = 50.8 mm and D = 31.7 mm galloping detonations are not discovered in both unstable CH_4 and C_3H_8 mixtures.

It is also worth noting that past galloping detonations in $CH_4 + 2O_2$ mixtures in the D = 1.5 and 3.2 mm diameter tube, the detonation velocity drops to a value of around ~0.6 V_{CJ} (see Fig. 6). The low-velocity detonation is maintained for long distances of propagation.

For stoichiometric $C_2H_2 + 5N_2O$ mixtures, similar behaviors as in $C_3H_8 + 5O_2$ and $CH_4 + 2O_2$ mixtures are observed indicating that mixtures with N_2O as oxidizer are unstable (see Fig. 7). In large tube diameters (i.e., D = 50.8 and 31.7 mm), no galloping detonations are observed in the $C_2H_2 + 5N_2O$ mixture. However, galloping detonations are observed in the three smaller tube diameters of D = 12.7, 3.2 and 1.5 mm. The near-limit behavior in $C_2H_2 + 5N_2O$ mixture is similar to that of $C_3H_8 + 5O_2$ and $CH_4 + 2O_2$.

Near the limits, smoked foils were used to determine the cellular detonation structure. Figure 8 shows examples of the smoked foil records in 12.7 mm tube when conditions are well within the detonation limits. The difference between the regularity of the cell pattern for stable mixtures (e.g., $C_2H_2 + 2.5O_2$ with 70% and 85% argon dilution) and unstable mixtures (e.g., $CH_4 + 2O_2$, $C_3H_8 + 5O_2$, $C_2H_2 + 5N_2O$) is clearly illustrated.

As the detonation limits are approached, the detonation decays from a multi-headed to single-headed spinning detonation. This is illustrated in Fig. 9. Upon entering the test section, the detonation is slightly overdriven with a corresponding multi-headed structure. It decays as it propagates, and near the end of the tube, the detonation decays to a single-headed spinning detonation. Note that the detonation velocity remains fairly constant throughout even though the structure changed from multi-headed to single-headed spinning detonation.

In the unstable $C_3H_8 + 5O_2$ mixture, a similar decay from an initially multi-headed detonation to a single-headed spinning wave is shown in Fig. 10. The cell pattern is more

irregular in this case than the stable mixture of Fig. 9. Again the detonation velocity remains fairly constant.

Smoked foil record of a galloping detonation in $CH_4 + 2O_2$ in the D = 12.7mm diameter tube is shown in Fig. 11. The decay to a single-headed spin is followed by the absence of any cell structure during the low velocity phase of the galloping cycle. The rapid acceleration from the low velocity phase back to an overdriven detonation to start the next cycle is indicated by the "abrupt" formation of cell structure. During the overdriven phase of the galloping detonation, a fine scale multi-headed structure can be observed. Therefore, periodic growth and decay of instability at the front occurs in a galloping cycle. These observations agree also with the results reported in [8]. For a stable mixture of $C_2H_2 + 2.5O_2 + 85\%Ar$ in the 12.7 mm diameter tube where no galloping detonations are observed, the smoked foil of Fig. 12 shows the decay of the single-headed spin and then the disappearance of cell structure past the single-headed spin.

4 Concluding remarks

Extensive investigations of the unstable velocity fluctuation and the variation of cellular detonation structure at the detonation limit of five explosive mixtures in different diameter tubes have yielded the following general conclusions:

- Away from the limits, the detonation propagates at a steady average velocity. The fluctuation of the local velocity is relatively small. The average velocity varies between $0.8V_{CJ} \leq V \leq V_{CJ}$ and in general, the velocity deficit increases with decreasing tube diameter.

- For stable mixtures of $C_2H_2 + 2.5O_2$ diluted with 70%Ar and 85%Ar, at the limit the detonation decays to a "low-velocity" detonation of $V/V_{CJ} \approx 0.4$ past the single-headed spin. The low-velocity detonation can propagate for long distances at a fairly constant velocity. Further reduction in initial pressure results in the failure of the low-velocity detonation to a flame.
- For unstable mixtures of $CH_4 + 2O_2$, $C_3H_8 + 5O_2$, $C_2H_2 + 5N_2O$, galloping detonations are observed in small diameter tubes (e.g., D = 12.7, 3.2 and 1.5mm). Numerous cycles of galloping detonations can be observed. The length of a galloping cycle is typically three hundred tube diameters and is not too sensitive to mixtures nor tube diameters. In larger diameter tubes, no galloping detonations are observed in the present investigation. However, narrower pressure range and longer test length for these diameter tubes may be required in the future work to verify if any galloping detonation exists at these conditions.
- In stable, highly argon-diluted mixtures no galloping detonations were observed even in small diameter tubes of D = 3.2 and 1.5 mm. The detonation just failed at the limit without any significant velocity fluctuation. No re-initiation of detonation was observed in the rest of tube. Hence it appears that galloping detonations are characteristics of "unstable" mixtures with irregular cell pattern since no galloping detonations were observed in stable mixtures.
- For low-velocity detonations, with $V/V_{CJ} \approx 0.4$, the detonation front has no cellular structure. Also, in the low velocity phase of a galloping cycle past single-headed spin, the detonation front also has no cellular structure. Spontaneous growth to a multi-headed

cellular front is observed as the detonation accelerates to an overdriven detonation to begin the next galloping cycle. Thus periodic growth and decay of instability at the front occurs in a galloping cycle. The cyclic behavior of galloping detonation is fairly reproducible which perhaps suggests that galloping detonation is more of a gas dynamic phenomenon of non-linear coupling between chemical reactions and the gas dynamic flow field.

Finally, since galloping detonations are not universally observed as the limit is approached for all tube diameters and mixtures used in this study, it appears that single-headed spin (the lowest transverse unstable mode) remains a reasonable criterion to define the detonation limit. However, there is a range of pressure in which single-headed spinning occurs. It is also difficult to determine precisely the onset of single-headed spin. Hence, defining detonation limits based on single-headed spin is at least an approximate criterion. Note that the detonation velocity cannot be used to define the limits since even at the limits prior to failure the detonation velocity seldom fails below 80% of the CJ value. The present study reinforces the suggestion that stability of the detonation front is responsible the propagation of the detonation wave.

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Figures captions

Fig. 1. A schematic of the experimental apparatus and a sample signals from the optical detectors.

Fig. 2. Velocity results for the $C_2H_2 + 2.5O_2 + 70\%Ar$ and $C_2H_2 + 2.5O_2 + 85\%Ar$ mixtures.

Fig. 3. Velocity results for the $C_2H_2 + 2.5O_2 + 70\%Ar$ mixture obtained using an extended tube.

Fig. 4. Velocity results for the $C_3H_8 + 5O_2$ mixture.

Fig. 5. Velocity results for the $CH_4 + 2O_2$ mixture.

Fig. 6. Low-velocity detonation for the $CH_4 + 2O_2$ mixture in 1.5 and 3.2 mm diameter tubes.

Fig. 7. Velocity results for the $C_2H_2 + 5N_2O$ mixture.

Fig. 8. Typical smoked foil records for both stable and unstable mixtures at the conditions well within the limit.

Fig. 9. Smoked foil records for the stable mixture at the conditions near the limit.

Fig. 10. Smoked foil records for the unstable mixture at the conditions near the limit.

Fig. 11. Smoked foil records for the unstable mixture at the conditions outside the limit.

Fig. 12. Smoked foil records for the stable mixture at the conditions outside the limit.

Figures

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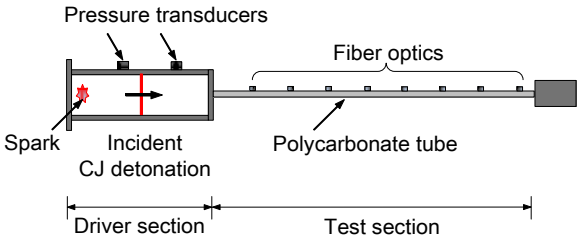
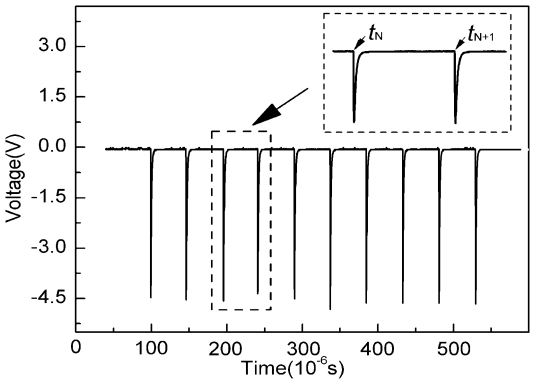


(a)



(b)

Fig. 1. a) A schematic of the experimental apparatus and b) sample signals from the optical detectors

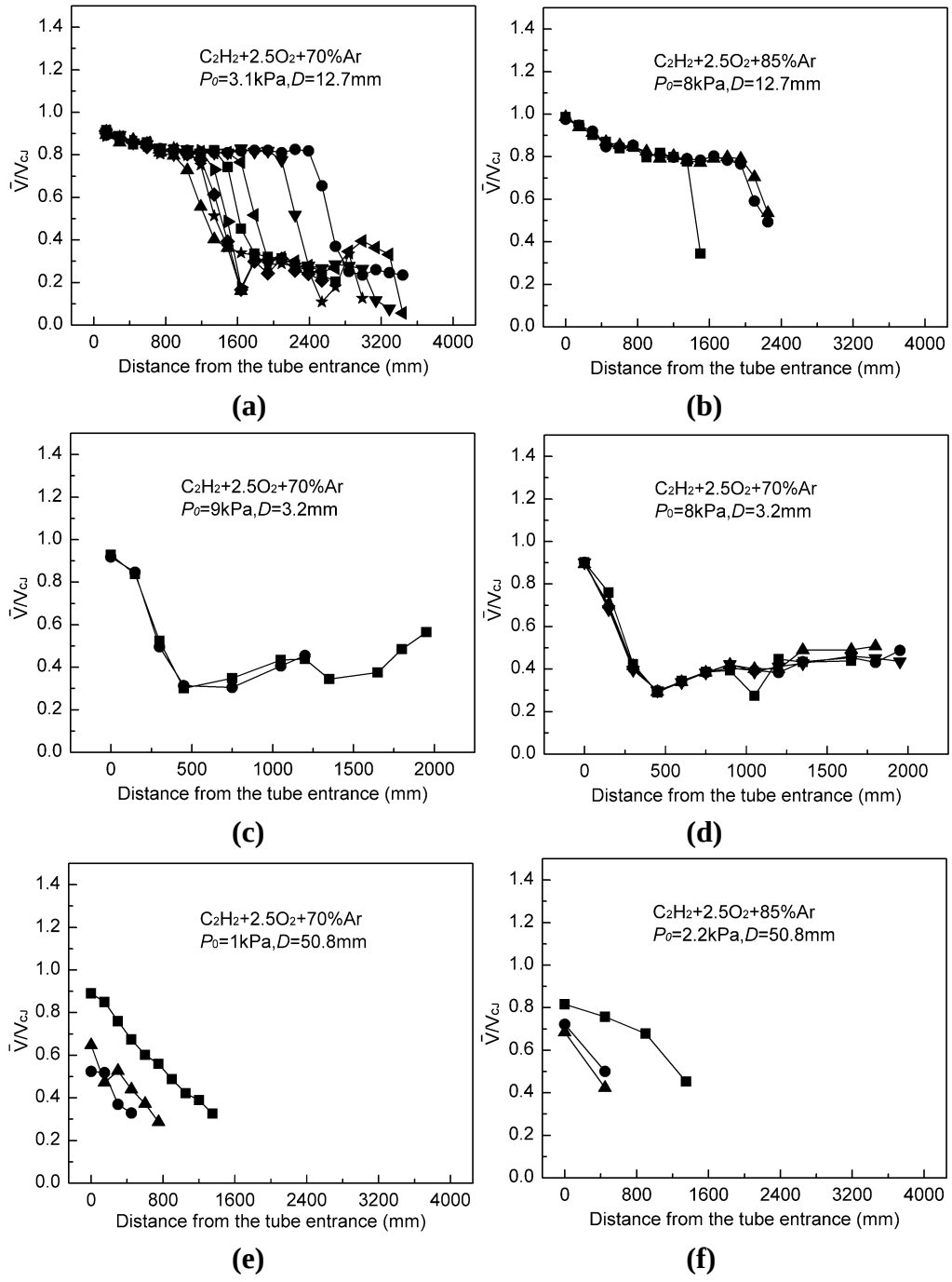


Fig. 2. Velocity results for the C_2H_2+2.5O_2+70%Ar and C_2H_2+2.5O_2+85%Ar mixtures

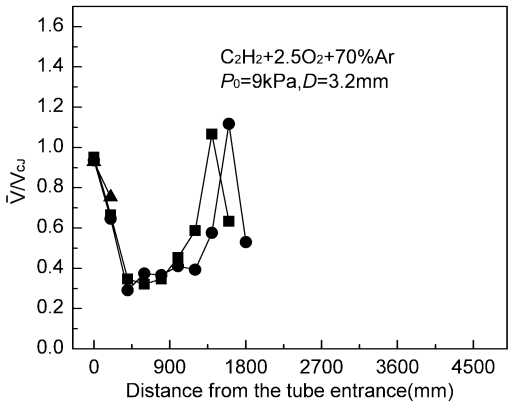


Fig. 3. Velocity results for the C_2H_2+2.5O_2+70%Ar mixture using an extended tube

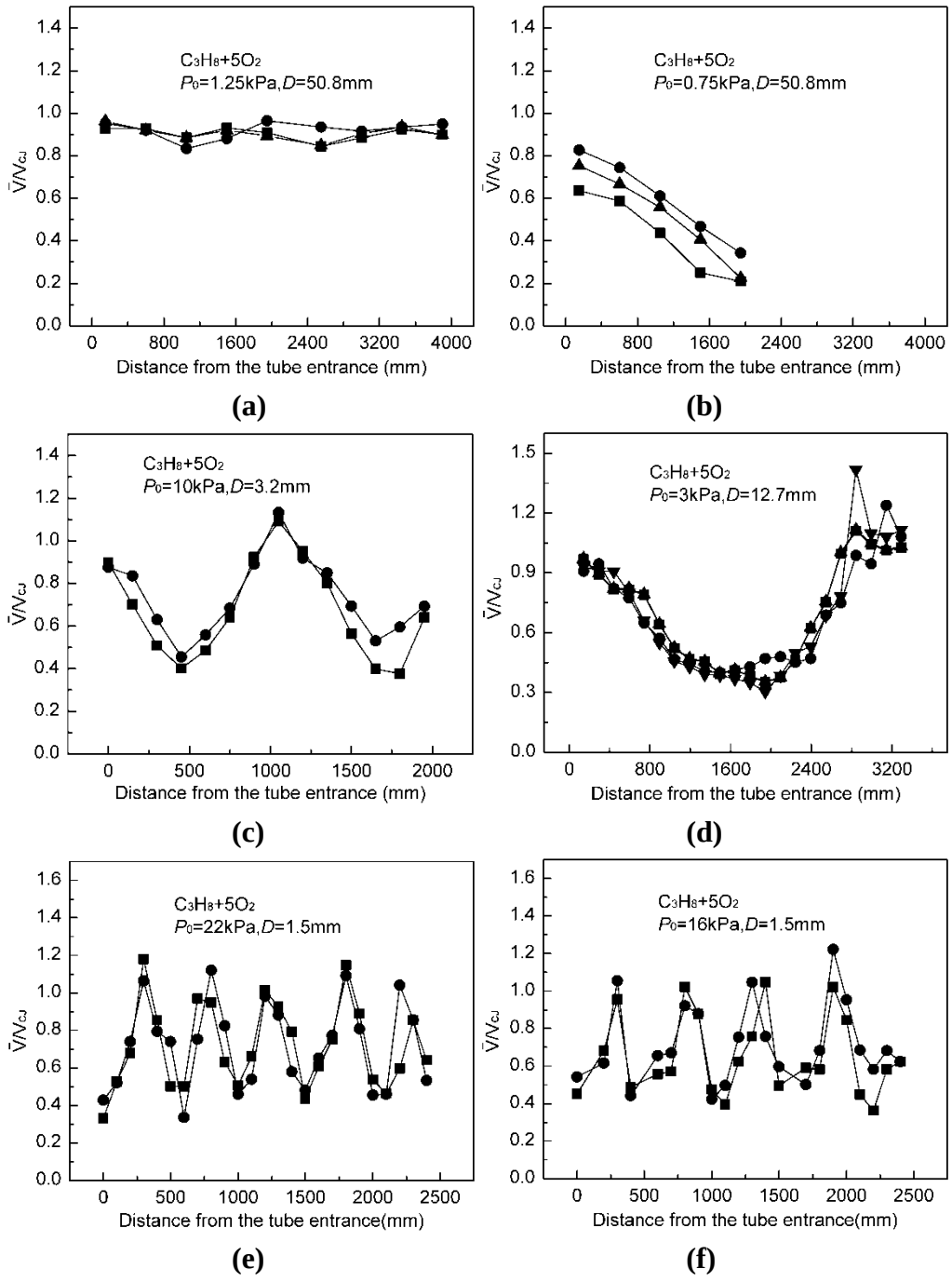


Fig. 4. Velocity results for the C_3H_8+$5O_2$ mixture

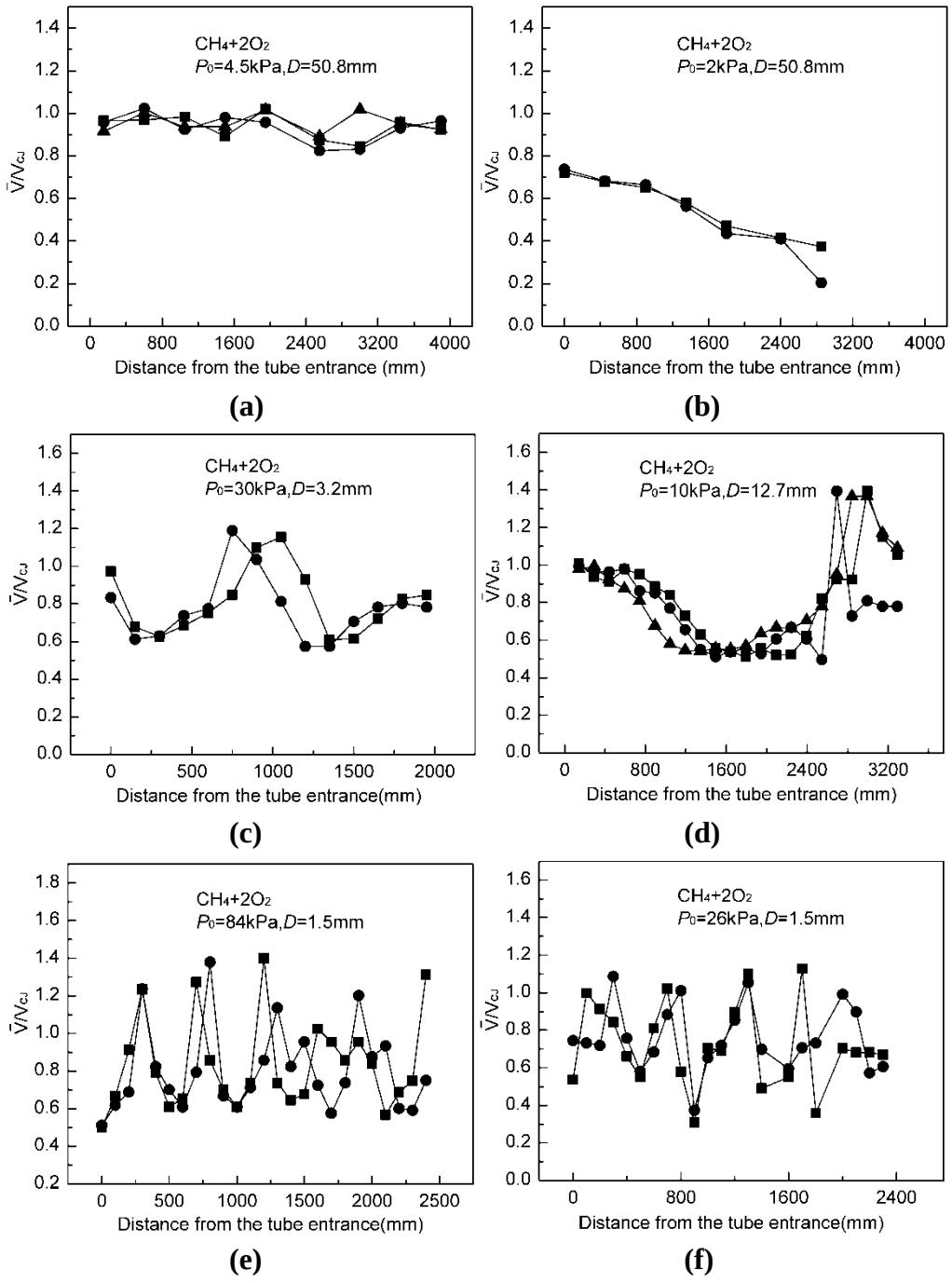


Fig. 5. Velocity results for the CH_4+$2O_2$ mixture

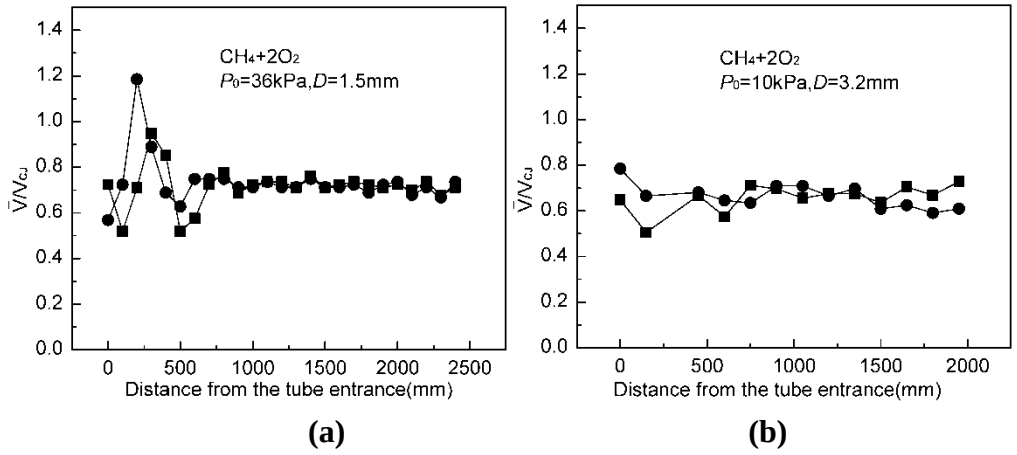


Fig. 6. Low-velocity detonation for the CH_4+$2O_2$ mixture in 1.5 and 3.2 mm diameter tubes

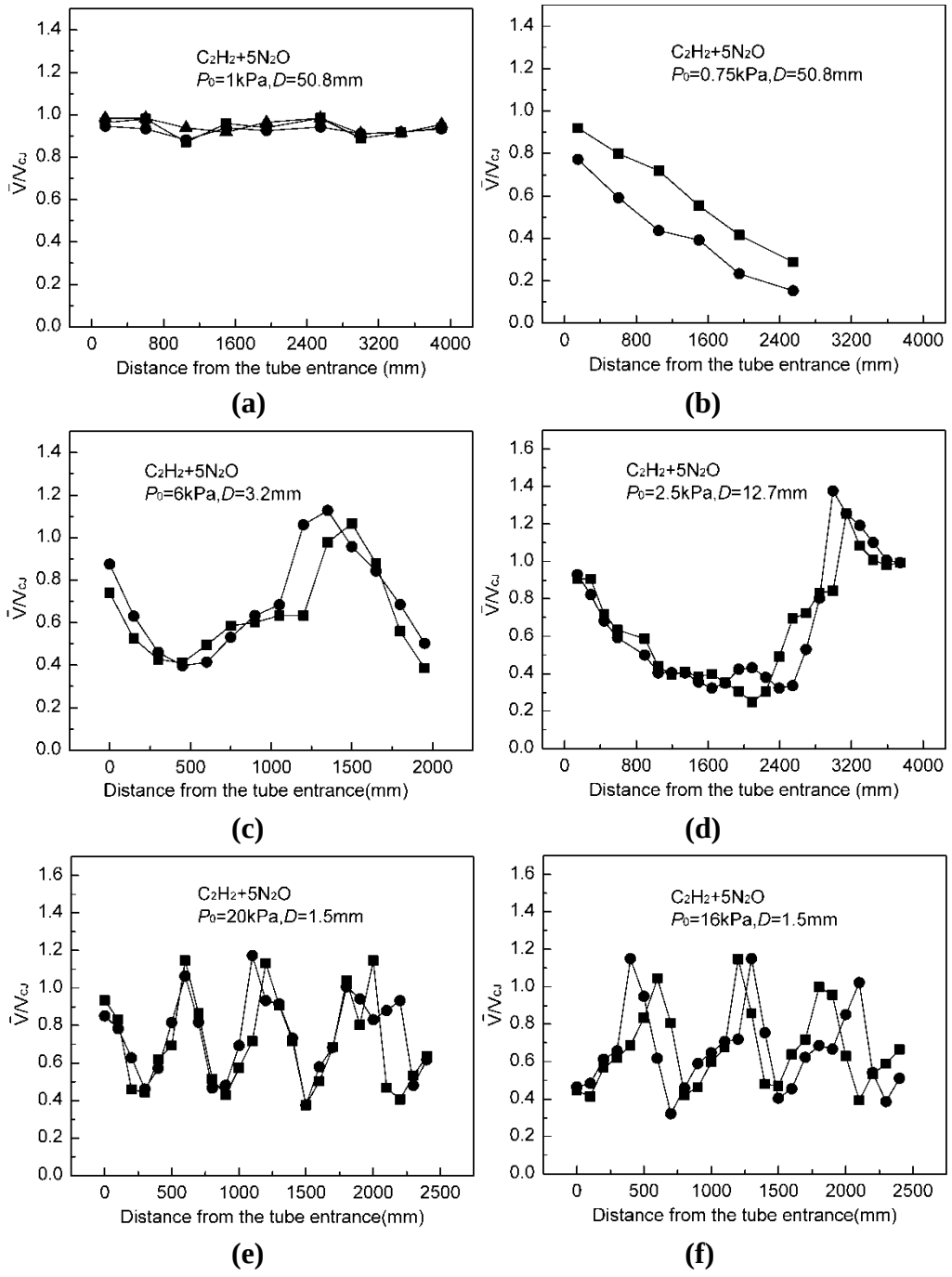


Fig. 7. Velocity results for the C_2H_2+5N_2O mixture

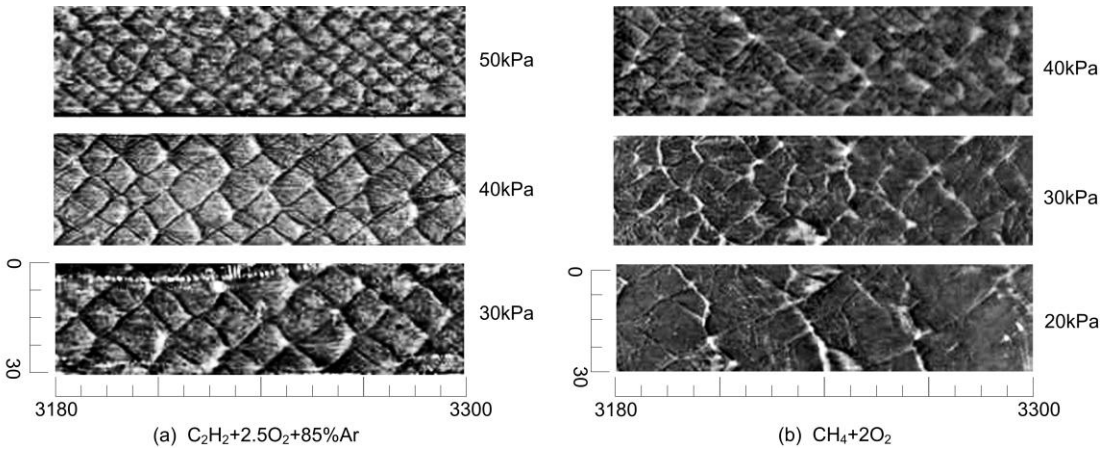


Fig. 8. Typical smoked foils records for both stable and unstable mixtures at the conditions well within the limits

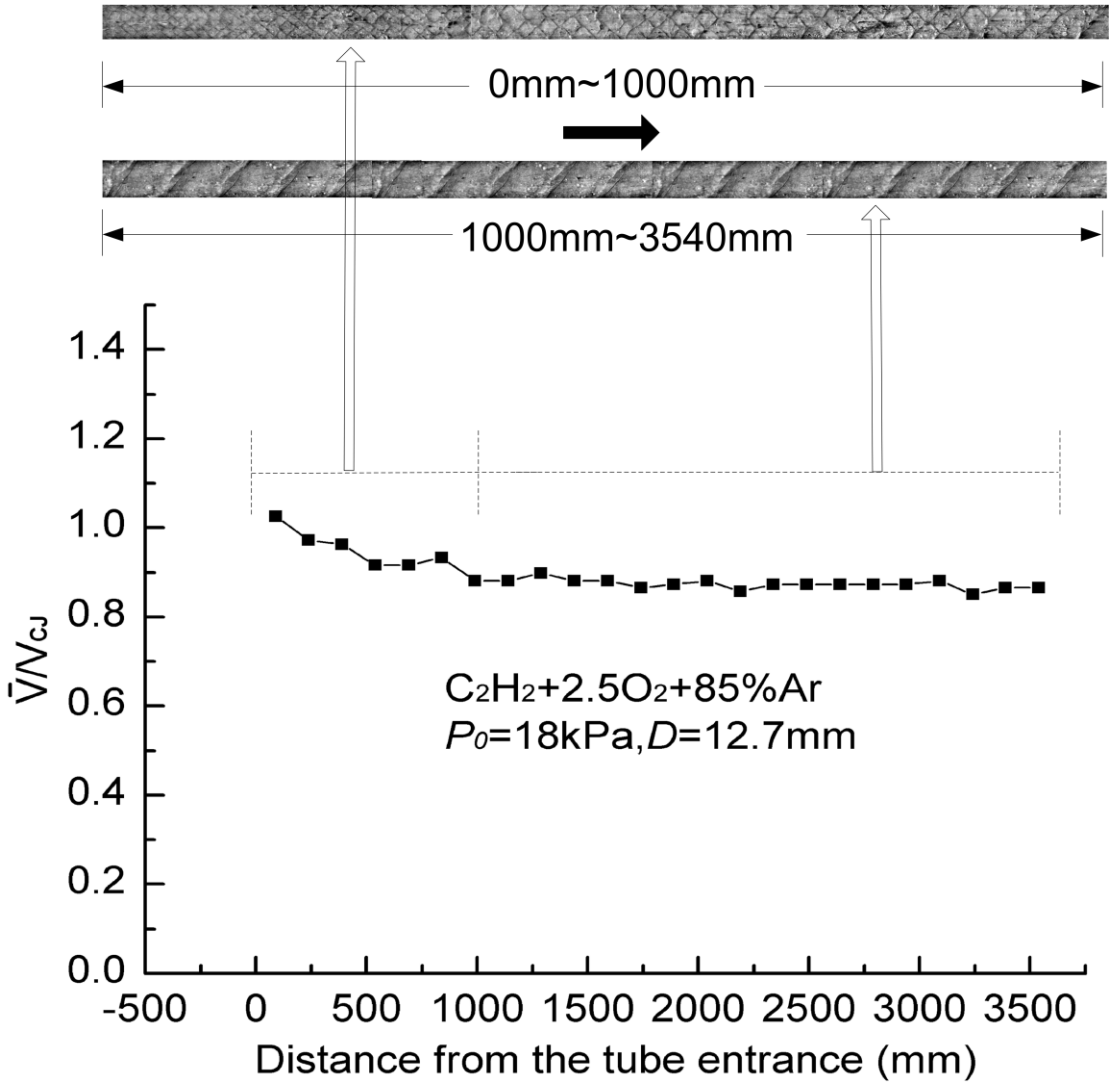


Fig. 9. Smoked foils records for the stable mixture at the conditions near the limit

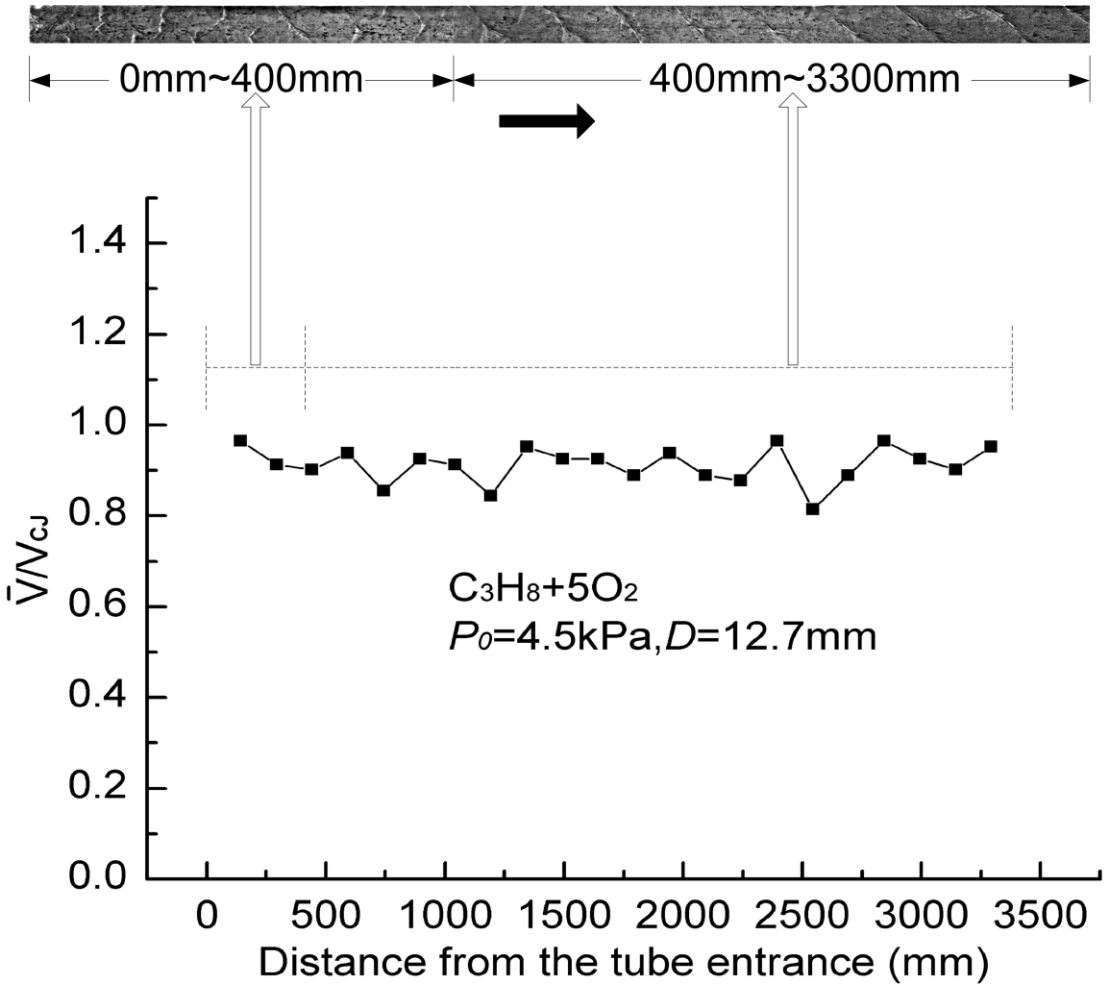


Fig. 10. Smoked foils records for the unstable mixture at the conditions near the limit

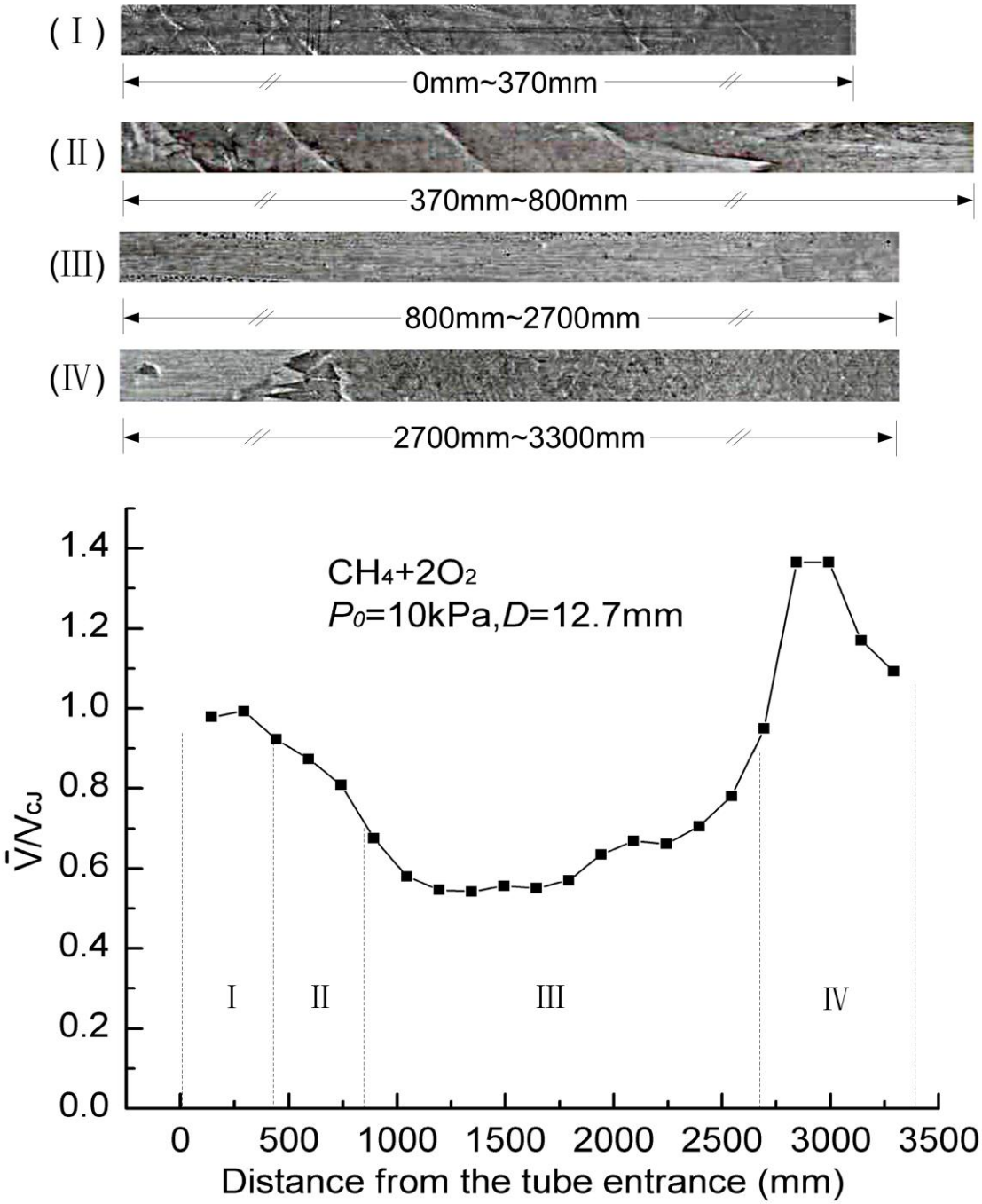


Fig. 11. Smoked foils records for the unstable mixtures at the conditions outside the limit

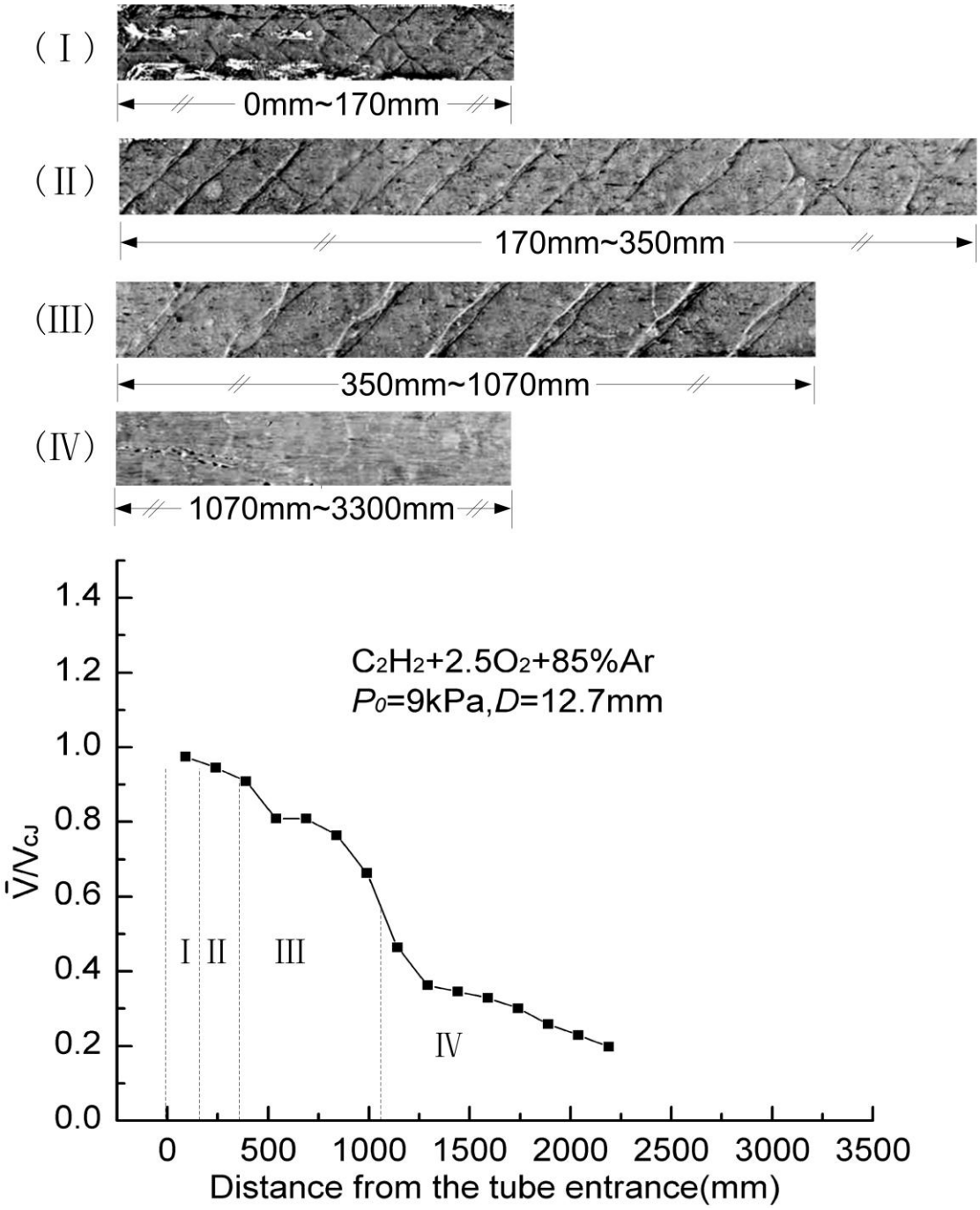


Fig. 12. Smoked foils records for the stable mixture at the conditions outside the limit