A COMPARATIVE STUDY OF DETONABILITY AND PROPENSITY TO SUSTAIN HIGH-SPEED TURBULENT DEFLAGRATIONS IN HYDROGEN AND METHANE MIXTURES

Maley, L.[1], Bhattacharjee, R.[1] and Radulescu, M.I.[1]
[1] Department of Mechanical Engineering, University of Ottawa, 770 King Edward Ave., Ottawa, K1N 6N5, Canada, LMaley@uottawa.ca

ABSTRACT

We've studied the conditions enabling a detonation to be quenched when interacting with an obstruction and the propensity for establishing subsequent fast-flame. Oxy-hydrogen detonations were found quench more easily than oxy-methane detonations, when comparing the ratio of gap size and the detonation cell size. High-speed turbulent deflagrations that re-accelerate back to a detonation were only observed in methane-oxygen mixtures. Separate hot-spot ignition calculations revealed that the higher detonability of methane correlates with its stronger propensity to develop localized hot-spots. The results suggest that fast-flames are more difficult to form in hydrogen than in methane mixtures.

1.0 INTRODUCTION

The aim of the current study is to examine the propensity of hydrogen and methane mixtures to maintain so-called choked, or fast flame propagation. These flames are believed to be the final stage of the deflagration to detonation transition (DDT) process. They are comprised of a shock wave and deflagration complex propagating quasi-steadily at approximately half the Chapman-Jouguet (CJ) velocity. These final stages of DDT are still poorly understood, sometimes even labeled as 'strange waves' [1]. For current turbulent flame theory to explain such fast-flames, burning velocities of several hundred m/s are required behind the leading shock in order to explain this behavior. It is difficult to explain these burning rates with current models. It is believed that these fast deflagration velocities are maintained via the action of pressure waves within this deflagration complex [2]. Thomas and co-workers [1] have conducted experiments studying this interaction of shock waves with flames and have shown that pressure wave interaction can cause the flame to become distorted and significantly increase the burning rates. Ciccarelli et al also observed such fast deflagration velocities in the presence of transverse pressure waves (see [2] and references therein). When Chao [3] investigated the fast deflagrations established downstream of a porous plate, a complex gas dynamic system which slowly accelerated towards CJ was observed. Radulescu and Maxwell [4] have studied the re-initiation of detonation waves following the wave interaction with a porous medium comprised of a two-dimensional array of staggered cylinders. From these experiments and the simultaneous numerical simulations**,** they reported that high-speed deflagrations require auto-ignition spots via shock compressions in order to drive strong pressure wave activity. This suggests that the high-speed deflagrations and unstable detonations may share the same mechanism of propagation. It has also been seen that unstable mixtures have a higher propensity to undergo DDT [5]. By studying the fast-flame, and hence the final stage of the DDT process, we are able to gain much insight into the process and the propensity of the fuel mixture to initiate a detonation.

The aim of the current study is to examine the propensity of hydrogen mixtures to undergo DDT experimentally starting with well-controlled initial conditions. To this end, this study examines the flow field of oxy-hydrogen and oxy-methane detonations in the wake of a single cylindrical obstacle. Bhattacharjee et al. [6] have determined that for sufficiently insensitive mixtures, the detonation is quenched as it diffracts around the obstacles, giving rise to a decoupled shock flame structure. In the present work, we study the dynamics of the re-initiation process of this decoupled wave. This experiment, using a single obstacle, allows the observation of an isolated approximate unit cell of the turbulent deflagration structure studied by previous investigators for the interaction of a detonation with a perforated plate ([3], [7]).

2.0 EXPERIMENTAL SET-UP

The experiments were performed in a 3.1 m long rectangular channel with a thin aspect ratio, 19.1 mm by 203.2 mm. The thin aspect ratio allows for the observation of quasi two-dimensional phenomena. The final section of the channel has glass walls allowing for optical access of the phenomenon, as seen in Fig.1. In order to visualize the phenomenon, three types of flow visualization have been used with a high-speed camera. A Z-type Schlieren photographic system has been implemented allowing for a visual field of 317.5 mm diameter. A direct shadowgraph system allowing for a visual field of 1 m (for more information see [8]) has been implemented, as well as the implementation of direct chemi-luminous photographs. The high-speed camera used in each visualization system was the Phantom v1210 high-speed camera which was used at a maximum rate of 140 000 frames per second. To illuminate the visual field**,** a 1600W Newport Arc Lamp was used in each system.

Figure 1 An illustration of the experimental channel

The test mixture used was stoichiometric oxy-hydrogen and oxy-methane mixed using the method of partial pressures and left to diffuse for a minimum of 24 hours before testing. The channel was evacuated to a pressure below 70 Pa before being filled with the gas to be tested.

The initial detonation waves were ignited using a capacitor discharge at one end of the tube opposite the view field. The detonations reached the CJ velocity before interacting with the obstacle. The experiments consisted of passing this self-sustained detonation around a cylindrical obstacle, 152.4 mm in diameter. For sufficiently low pressure, the diffraction of the wave around the downstream side of the cylinder causes the detonation to fail, decoupling the shock wave and reaction zone. The resulting dynamics of the fast-flame structure were monitored with time-resolved Schlieren, shadowgraph, and chemi-luminescence photographs.

The Schlieren and shadowgraph images have been modified to enhance the features present in the images. A background subtraction has been done performed, this is courtesy of S. She-Ming Lau-Chapdelaine and the contrast has been modified for each image to allow the important characteristics to be viewed more easily.

3.0 RESULTS

Fig. 2 shows an example of a detonation diffraction and re-initiation around the obstacle in hydrogen-oxygen. The shadowgraph permits to identify regions of density gradients, while the self-luminosity permits to identify clearly detonation combustion. As the detonation wave propagates around the obstacle, the shock and reaction zone locally decouple. For these conditions, however, the reflection of the two diffracted shocks permits the detonation to re-establish. While this transient has been addressed in detail by Bhattacharjee et al. [6], the present study focuses on less sensitive mixtures, where the prompt re-initiation showed in Fig. 2 does not occur. The critical pressure for the detonation re-initiation around the obstacle was found to be 14.4 kPa for oxy-hydrogen and 13.8 kPa for oxy-methane. It is customary to report the limits in terms of characteristic cell size λ in relation to the gap size b, which is 51 mm in the present case. This yields a ratio of $b/\lambda = 6.5$ for the hydrogen mixture and a value of $b/\lambda = 1.5$ for the methane mixture. The lower ratio in methane-oxygen suggests that, for a fixed cell size, the mixture is more likely to initiate a detonation than in hydrogen-oxygen.

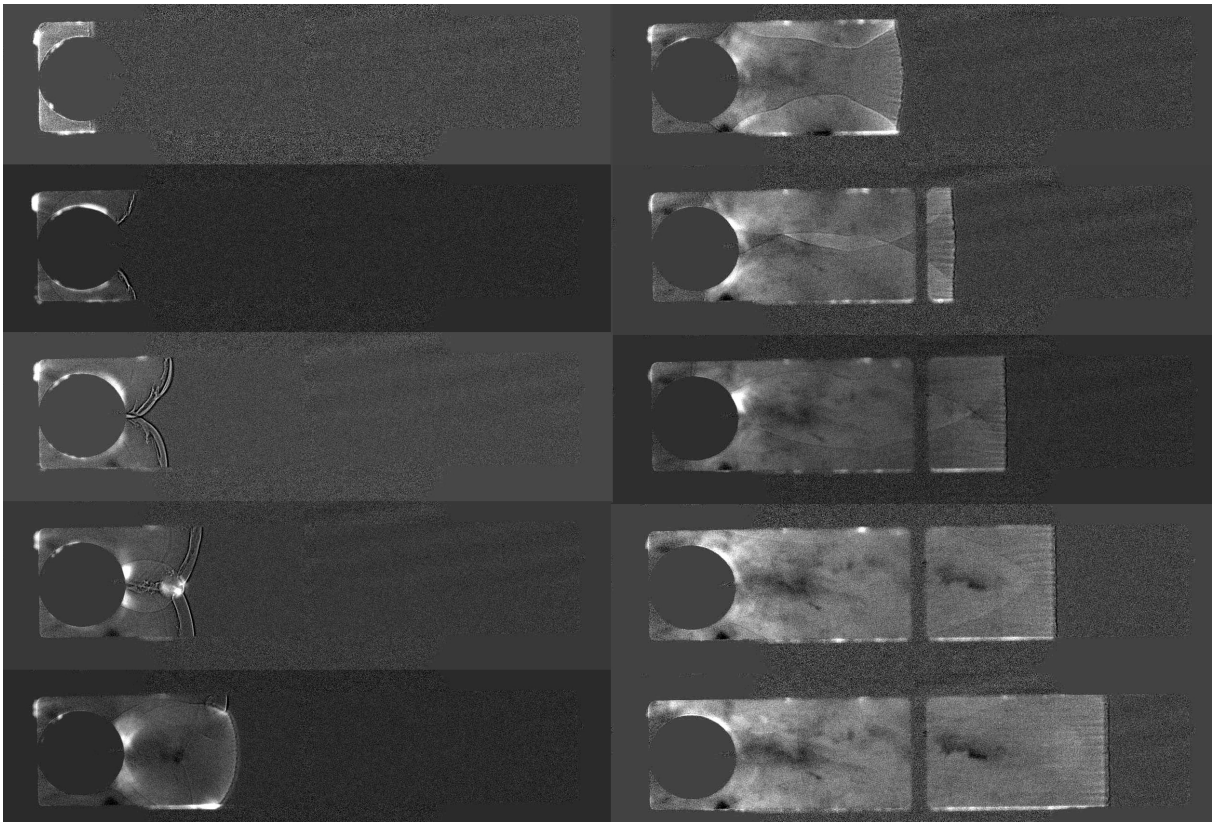

Figure 2 A composite image of oxy-hydrogen transition to detonation during the transitional Mach reflection at an initial pressure of 14.47kPa

For the hydrogen mixtures, experiments conducted at pressures less than the critical pressure, did not reveal the presence of any fast flame. Fig. 3 shows an example of the resulting decoupled shock and reaction zones, where the flame trails behind the leading shock at a growing distance. Under these conditions, the burning rate of the flame is not able to keep up to the shock wave traveling at Mach 3.13 after the Mach reflection, decaying to Mach 1.87 by the last frame. Following the reflection, the reaction zone remains fairly smooth and no major turbulent structures are present.

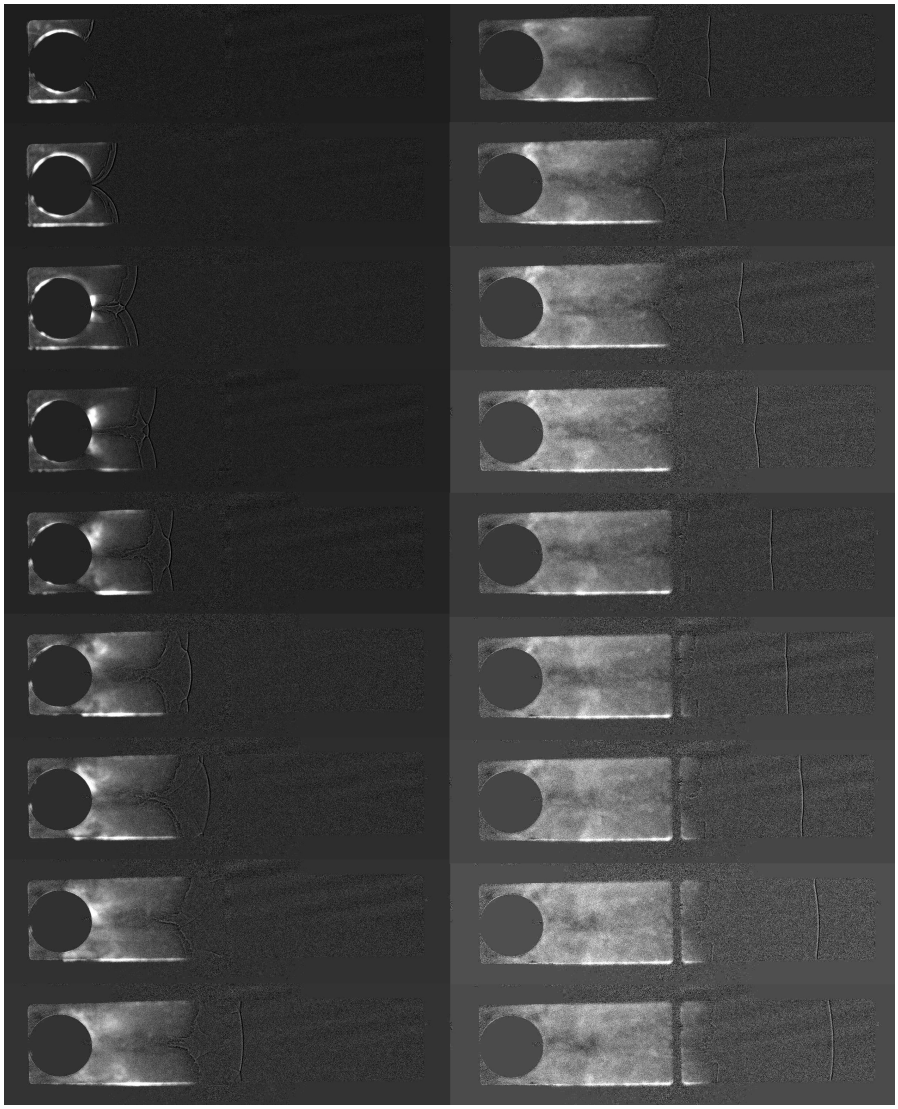

Figure 3 A series shadowgraph images, with 34.91 microsecond a inter-frame time, of a de-coupled shock flame coupled in oxy-hydrogen at 8.61kPa initial pressure

The experiments in methane-oxygen, however, revealed a significantly different picture. Fast flames, punctuated by the presence of hot spots were observed. Fig. 4 shows a typical example of a hot spot created from the reflection of the attenuating diffracting shocks. This image shows the dynamic evolution of the wave in the near field and the development of the high-speed deflagration known as the fast-flame. The origin of the first hot-spot is due to the jet entrained along the axis of symmetry. The resulting violent pressure evolution generates a series of transversely propagating shock waves, which then interact with the burned/unburned interfaces, locally amplifying the burning rates. This transient has been documented in our previous works ([6], [7]) both experimentally and numerically. Interestingly however, this localized explosion does not lead directly to a detonation.

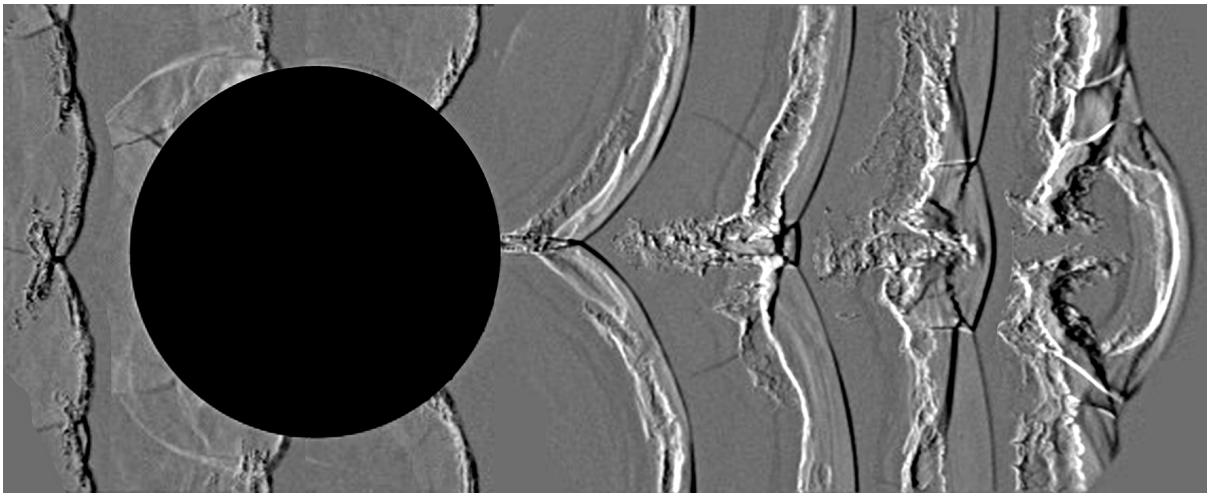

Figure 4 Composite of Schlieren frames illustrating the diffraction of a detonation wave around the cylindrical obstacle and the local re-ignition after shock reflections due to jet behind the Mach stem obtained at 8.2 kPa initial pressure.

Fig. 5 shows the evolution of the flow field. Again, the Mach reflection in the wake of the obstacle is again strong enough to create a local hot-spot. This kernel of ignition, caused by the transitional Mach reflection of the leading shock, drives transverse pressure waves into the surroundings increasing the local burning rate. This injection of energy feeds the shock wave and in turn increases the local burning rate, seen in frames 6, 7, and 8. The transverse waves interact and reflect to form new generations of hot-spots as exemplified in frames 9 and 19. From the series of images in Fig. 5 it can be seen that with every subsequent transverse reflection a new generation of hot-spots are formed. This increased burning rate allows the flame to remain coupled to the shock and continue the propagation at approximately half the CJ velocity, accelerating slowly. Eventually, this fast flame transits to a detonation wave once one mode becomes sufficiently strong.

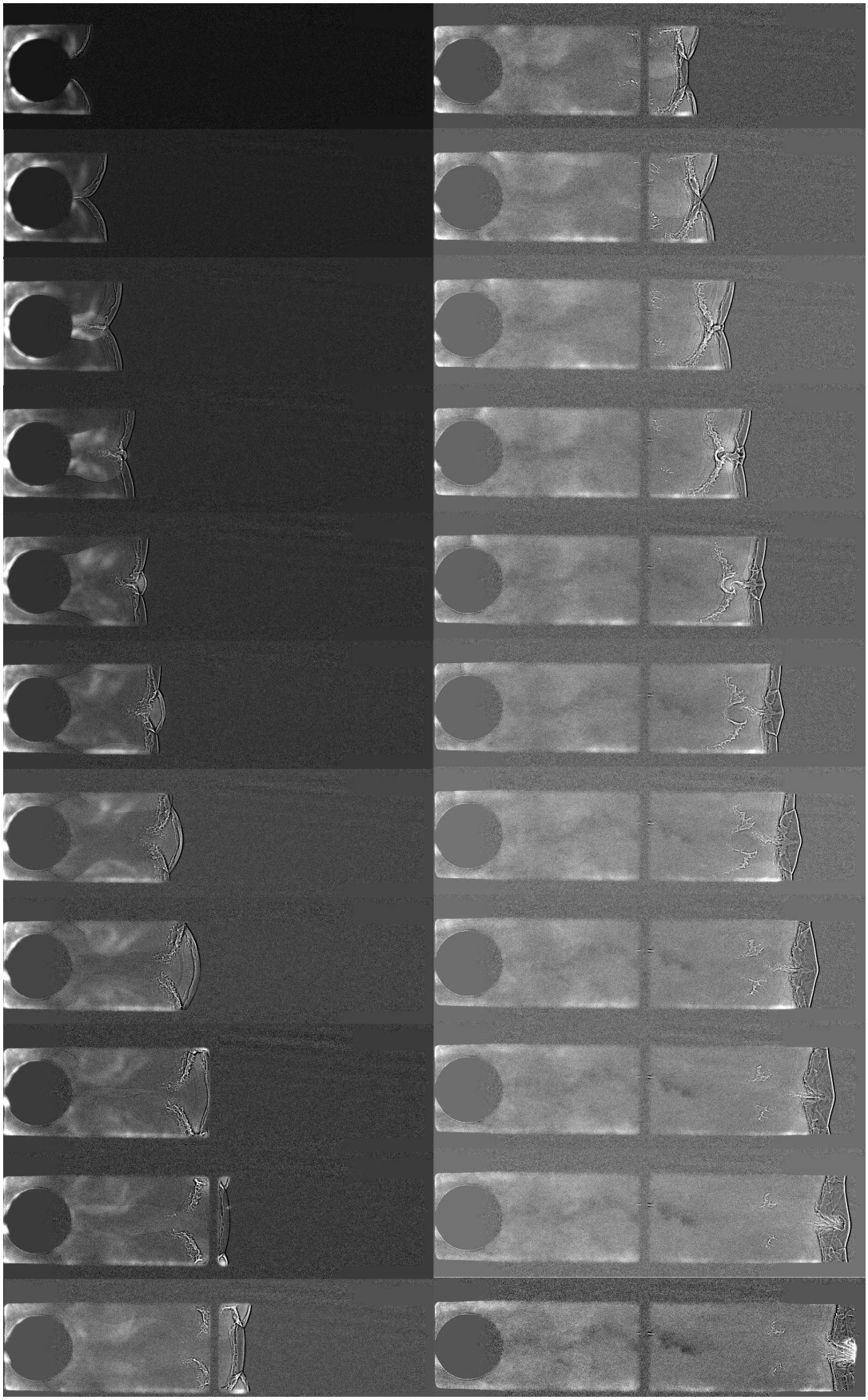

Figure 5 A series of shadowgraph images, with 25.63 microsecond an inter-frame time, of local hot-spot ignition leading to a fast flame structure at 8.48 kPa initial pressure in oxy-methane.

A transition to detonation within the field of view is shown in Fig. 7. This series of shadowgraph images shows a typical development of the fast-flame transition to detonation. From the initial creation, the fast-flame structure continues to accelerate and eventually result in the transition to detonation. Fig. 7 begins with the development of the fast-flame structure by the explosion kernel ignited by the Mach reflection, seen by frame 7. The transverse waves caused by this ignition spot increase the local burning rate, frames 7 and 8, until they reflect in frame 9. This transverse wave reflection results in a hot-spot that increases the local burning rate sufficiently for the flame to transit to detonation.

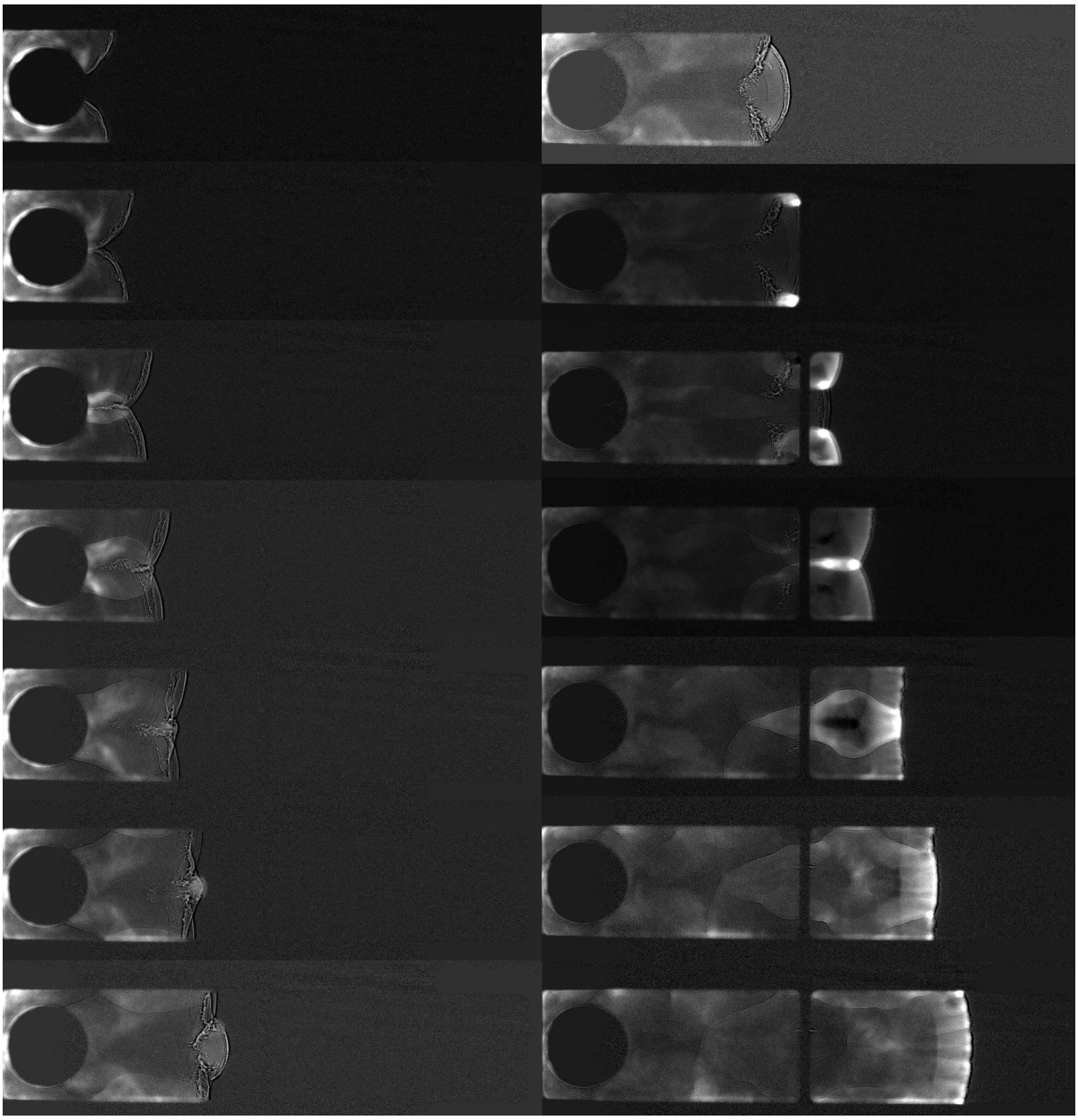

Figure 7 A composite shadowgraph image of a fast flame transit to detonation in oxy-methane at 11.03 kPa initial pressure.

The transition of the fast flame takes place when the transverse wave interaction is able to develop a hot-spot with a large sudden energy release. Fig. 8 shows the direct chemi-luminescence photograph of a similar transition. Using direct chemi-luminescence, we are able to better visualize the hot-spots.

Fig. 8 shows the same dynamics as Fig. 7, although the transition to detonation takes place further down the channel. In the 5th frame of Fig. 8, the transverse wave interactions give rise to another generation of hot-spots. These hot-spots continue the local amplification of the wave. By the 9th frame, the transverse wave interactions lead to a hot-spot with sufficient energy release for the wave to transit to detonation as seen in the 10th frame. This shows that the mechanism for propagation of the fast-flames, the transverse wave interactions leading to local explosion kernels, is the same as the propagation mechanism for unstable detonations. These local hot-spots provide sufficient energy release to maintain the propagation of the coupled fast-flame.

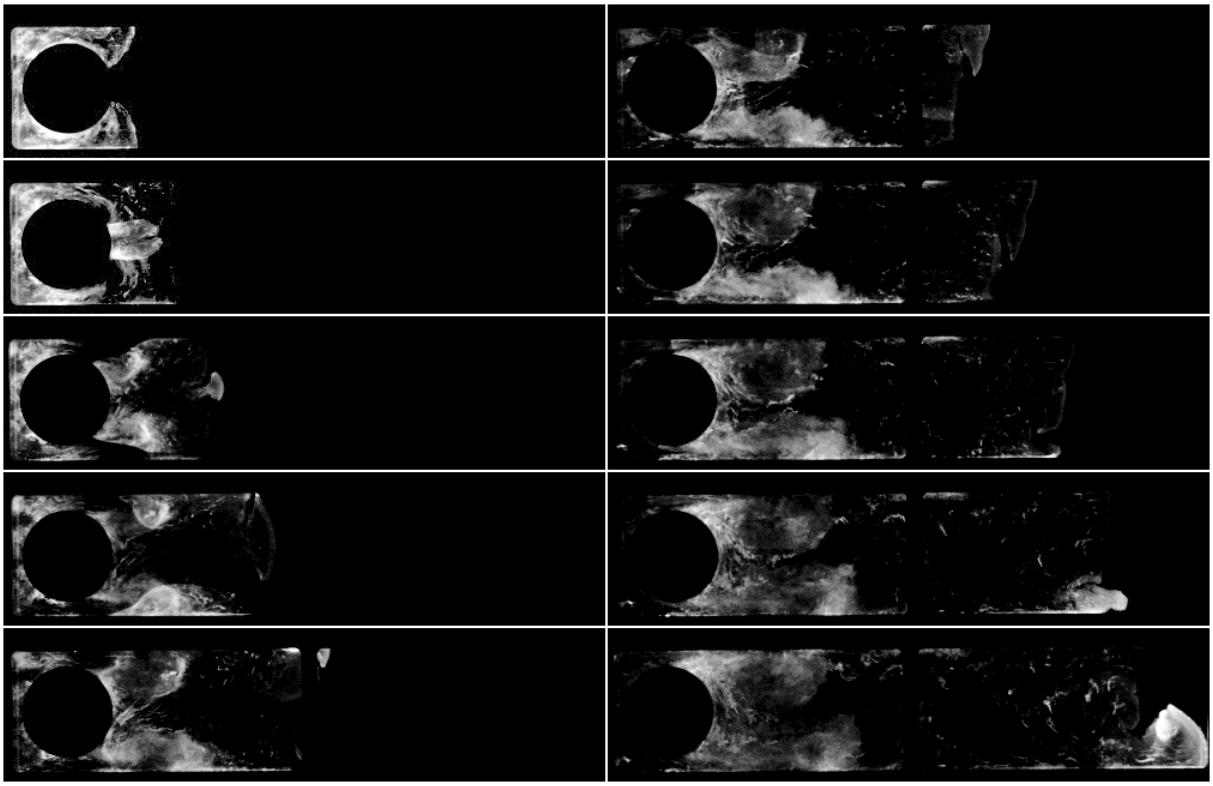

Figure 8 A composite image of a direct chemi-luminescence showing the structure of a fast-flame which eventually transitions to a detonation. Using a 1microsecond exposure, the inter-frames time is 57.1 microseconds

On the contrary, in hydrogen oxygen, this amplification is not observed. In some cases, we do observe an initial hot-spot from the first reflection, such as in Fig. 6. However, the hot-spot ignition does not lead to the same coupling effects of the shock flame complex and the development of the fast-flame structure is not observed. The subsequent decay of the shock flame structure results in a fully decoupled wave. As the shock flame complex propagates down the channel, the flame continues to lag further behind the leading shock and the shock speed decays. No flame acceleration is observed after the initial ignition kernel. The transverse waves are unable to increase the burning rate sufficiently in order for the energy release to sustain the flame speed required for the fast-flame structure.

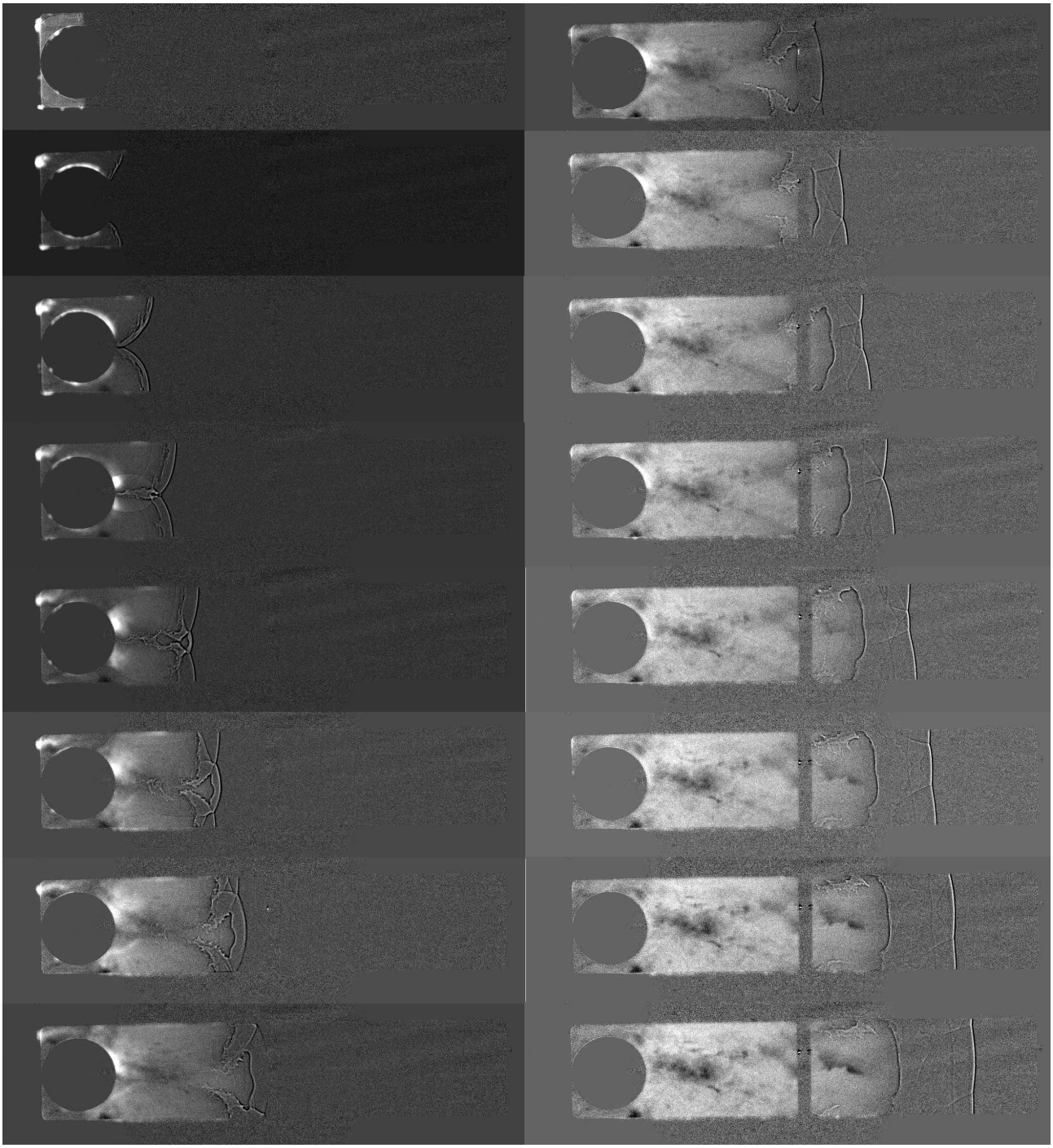

Figure 6 A composite of shadowgraph images of an ignition kernel and subsequent decay of the shock flame complex in oxy-hydrogen at 13.78kPa.

4.0 DISCUSSION

The wave dynamics demonstrated above suggest that the main mechanism for fast-flame propagation is the same propagation mechanism as unstable detonations; the increased burning rate caused by the transverse wave interactions leads to local explosions that continue to drive new transverse waves, each increasing the local burning rate and locally amplifying the wave. The underlying process for this behavior remains to be elucidated.

Fig. 9 shows the shock speed profile recorded in hydrogen-oxygen and methane-oxygen from the subsequent movie frames. For the two experiments, the speed is reported along the upper wall, centerline, and lower wall. For hydrogen oxygen, shown in Fig. 9a, the lead shock continuously decays and shows little fluctuations. For the methane-oxygen case, Fig. 9b, the fluctuations are much more important. These can be correlated with the discrete ignition of hot-spots from subsequent reflections. Also noteworthy in this case is the slow acceleration of the front.

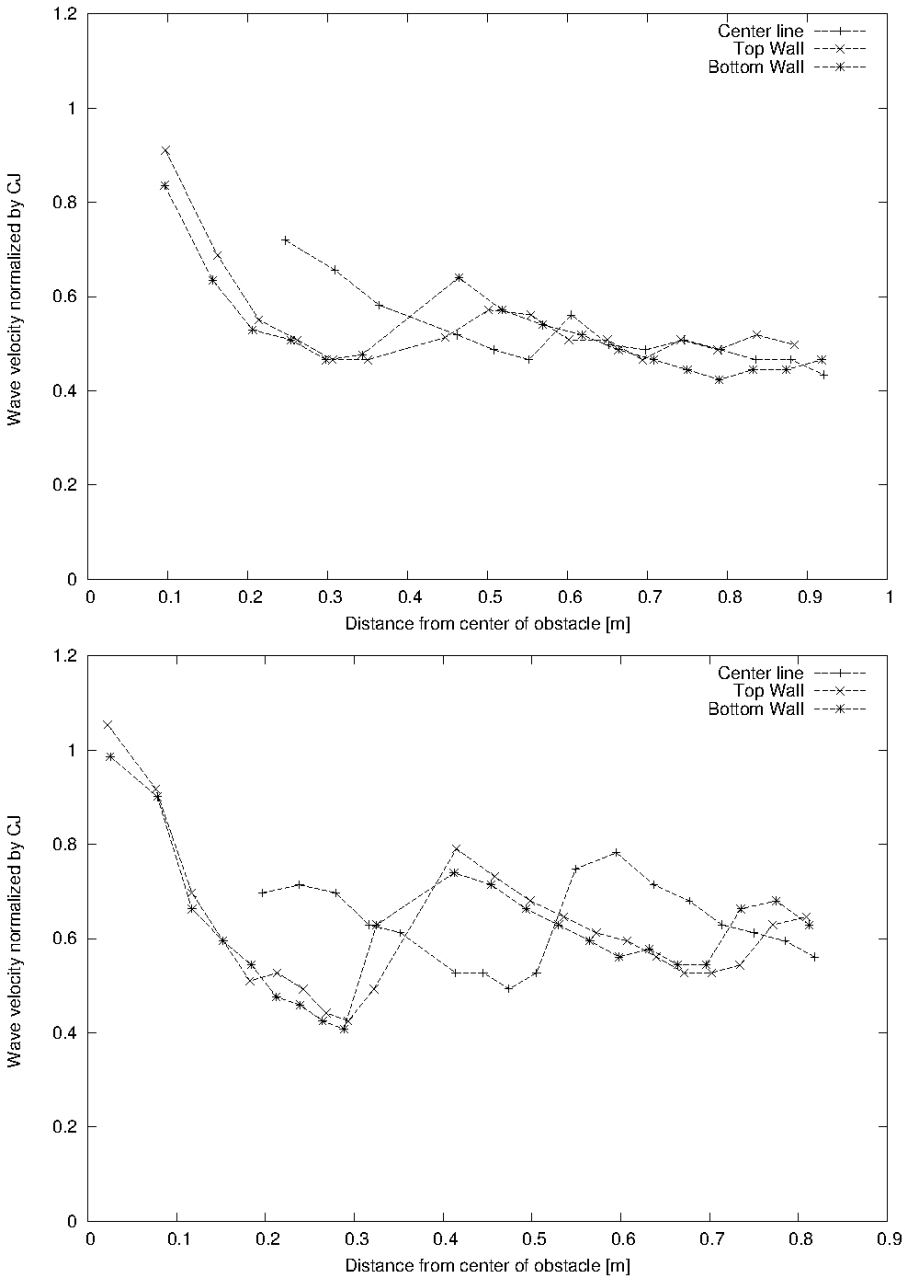

Figure 9 a) Oxy-hydrogen wave velocity normalized by the oxy-hydrogen CJ velocity vs. distance from centre of obstacle, corresponding to Fig. 6. b) Oxy-methane wave velocity normalized by the oxy-Methane CJ velocity vs. distance from centre of obstacle, corresponding to Fig. 5.

A potential explanation for the much stronger propensity of methane to amplify such hot-spots rests on differences in the thermo-kinetic parameters in the two fuels. Very recently, Radulescu, Sharpe and Bradley associated the likelihood of a mixture to develop hot-spots to a stability parameter χ [10].

$$\chi = \frac{t_i}{t_e}\frac{E_a}{RT}\frac{Q}{RT} \tag{1}$$

The parameter E_a / RT is the usual non-dimensional activation energy, T is the local temperature, R is the universal gas constant, Q / RT is the non-dimensional heat release, t_i / t_e is the ratio of induction to reaction times. Radulescu has demonstrated that a higher value of the χ -parameter indicates a higher propensity for hot spot development and propensity to detonate [10]. We evaluated this parameter taking a shock propagating at 70% of the CJ velocity as the characteristic speed (see Fig. 9). The χ -parameter yields 2×10^6 in hydrogen-oxygen and 8×10^6 in methane-oxygen. While this indicates that both mixtures are susceptible to hot-spot formation, the methane mixture has a stronger propensity. The difference of the values for these mixtures is, however, not sufficiently large to fully explain the different behaviour seen in the experiments.

The difference in behaviour cannot be rationalized on the magnitude of ignition delays. For example, taking again the state behind a shock propagating at 70% of the CJ value as the reference state, we obtain ignition delays of 5 ms in the hydrogen-oxygen mixture and 16 ms in the methane-oxygen mixture. The longer delay in methane would indicate the opposite trend then that observed experimentally. Nevertheless, both predictions are much too long to be relevant to the experiments, indicating the potential role of localized hot-spots and diffusive processes.

The fast-flames have been seen to consistently transition to detonation during a strong transverse wave reflection. Significant study has been done into the dynamics of Mach reflections and the effect on the combustion process. Previous works have explored the forward jetting effects caused by the Mach reflection process and the turbulent effects which are associated with this process [6], [7], [11], as shown in Fig. 4. Indeed, the higher value of adiabatic exponent in the hydrogen system of 1.36, compared to 1.2 in the methane system, is sufficient to supress the jetting effect [11]. This remains to be verified by detailed observations of the shock reflection process in hydrogen. A well-controlled experimental investigation is underway to isolate the shock reflection process, and investigate the influence of chemistry and the chemical kinetics on the gas dynamics[12].

5.0 CONCLUSION

The methane-oxygen system was shown to sustain fast-flames while the hydrogen system does not. The limits for hydrogen detonation transitions have been seen to be more strict than those of oxy-methane. The lower propensity for detonation is due to the lack of fast-flames, which eventually transit to detonation. The oxy-hydrogen detonation is more easily quenched once initiated, characterized by the larger b/λ ratio. Overall, it is more difficult to initiate a detonation in the oxy-hydrogen mixture, and that the corresponding detonations are more easily quenched once initiated.

The transverse shock waves were found to play a very important role in the propagation mechanism for the fast-flame structure. The experimental evidence points to a mechanism similar to unstable detonation waves, i.e., the turbulization of the reaction zone by the transverse pressure waves originating from hot-spots. The absence of such a regime in the hydrogen system is compatible with the trends expected for hot spot formation recently suggested by Radulescu et al. Nevertheless, the role of forward jetting that occurs during the Mach reflection process, and the turbulent mixing which is associated with this process, may play a significant role. Further investigation is required to fully clarify this process.

6.0 BIBLIOGRAPHY

1. Thomas, G., Bambrey, R., and Brown, C., Experimental observations of flame acceleration and trasition to detonation following shock-flame interaction, *Combustion Theory and Modelling*, vol. 5, no. 4, pp. 573–594.
2. Ciccarelli, G., Johansen, C., and Kellenberger, M., High-speed flames and DDT in very rough-walled channels, *Combustion and Flame*, vol. 160, no. 1, pp. 204–211, Jan. 2013.
3. Chao, J. C., Critical deflagration waves that lead to the onset of detonation, Ph.D. dissertation, McGill University, Montreal, Canada, 2006.
4. Radulescu, M., Maxwell, B. M., The mechanism of detonation attenuation by a porous medium and it's subsequent re-initiation, *J. Fluid Mech.*, vol. 667, pp. 96–134.
5. Kuznetsov, M.S., Alekseev, V.I., and Dorofeev, S.B., "Comparison of critical conditions for DDT in regular and irregular cellular detonation systems," Shock Waves.10:217-23 (2000).
6. Bhattacharjee, R., Lau-Chapdelaine, S. S. M., Maines, G., Maley, L., and Radulescu, M.I., Detonation re-initiation mechanism following Mach reflection of a quenched detonation, *Proceedings of the Combustion Institute*, vol. 34, 2013, pp.1893–1901.
7. Grondin, J.-S., Lee, J. H. S., Experimental observation of the onset of detonation downstream of a perforated plate, *Shock Waves*, vol. 20, no. 5, pp. 381–386, Oct. 2010.
8. Denis, K., Maley, L., Radulescu, M.I., Implementation of Large Scale Shadowgraphy in Hydrogen Safety Phenomena, *International Conference on Hydrogen Safety*, 2013.
9. Bhattacharjee, R., Experimental investigation of detonation re-initiation mechanisms following a Mach reflection of a quenched detonation, M.A.Sc. Thesis, University of Ottawa, Ottawa, Canada, 2013.
10. Radulescu, M.I., Sharpe, G. J., Bradley, D., A universal parameter quantifying explosion hazards, detonability and hot spot formation: the χ number, Proc. of the Seventh International Seminar on Fire & Explosion Hazards, 2013.
11. Mach, P., Radulescu, M. I., Mach reflection bifurcations as a mechanism of cell multiplication in gaseous detonations, *Proceedings of the Combustion Institute*, vol. 33, no. 2, pp. 2279–2285, 2011.
12. Maley, L., Armstrong, J., and Radulescu, M.I., Experimental Implementation of a Converging Diverging Nozzle Technique to Study Shock Reflections in Reactive Gases, *Proceedings of the* 23rd *International Colloquium on the Dynamics of Explosions and Reactive Systems*, 2011.