Propagation of Fast Deflagrations and Marginal Detonations in Hydrogen-Air-Additive Mixtures

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Introduction

After a possible failure of any hydrogen infrastructure at industrial or, in future, civil sites, freely propagating flame-fronts are very likely to arise due to both, the low required ignition-energy and the wide ignition-range ($4-75$ Vol.%) of hydrogen-air mixtures. The pressure-loading of the combustion depends strongly on its mode, either deflagrative or detonative. In case of a subsonic deflagration of a hydrogen-air mixture, a structure of a building can withstand the pressure rise of the combustion process. Although a direct ignition of a detonation is very unlikely due to the required high energy-source, an at first slow deflagration can turn into a detonation by a deflagration-to-detonation transition (DDT) process, which can endanger the integrity of the building structure. The understanding of this highly transient as well as complex physical transition process was mainly achieved by optical measuring techniques. Urtiew and Oppenheim [UO66] showed in their pioneering work by means of the classical Schlieren-Cinematography the possible modes of the transition process from a fast, turbulent deflagration into a detonation. Although this process is qualitatively quite well understood due to the extensive research in that area, there are still gaps in the understanding of the propagation mechanisms of fast deflagrations and detonations as well as their transition process, at least if the geometrical detonability limits (cf. [BCD$^+$00]) are concerned. Due to the progress in the development of new optical measuring techniques, a deeper insight into these highly transient combustion phenomena can now be achieved. This paper reports on results of fast deflagrations and detonations, obtained by conventional as well as sophisticated optical measuring techniques such as the color-schlieren technique and the planar laser-induced predissociation fluorescence, which facilitates the visualization of the OH-radical distribution in a very thin layer (≈ 0.3 mm). The experiments where conducted with an explosion tube (Ø 66 mm, length 6.5 m), which was closed at both ends. Flame acceleration was provided by means of a periodic distribution of obstacles with a different length L_{OP}, blockage-ratio BR, and obstacle spacing l_{SP} in the first part of the tube, in order to get a spectrum of flame-velocities in the optical accessible section in the middle of the tube with a length of 30 cm. Besides pure hydrogen-air mixtures at ambient temperature and pressure, the influence of additives was studied.

Results

Flame propagation in the obstacle path

The process of flame acceleration in the obstacle path is the basis for the investigation of deflagration-to-detonation transition in dependence of the flame velocity in the unblocked tube section. In Fig. 1, the maximum flame velocity in the obstacle-path in dependence of the applied obstacle-configuration as well as the equivalence ratio ϕ is shown. For the flame-propagation in obstacle-filled tubes, the criteria of Peraldi et al. [PKL86] are often used as a reference for the

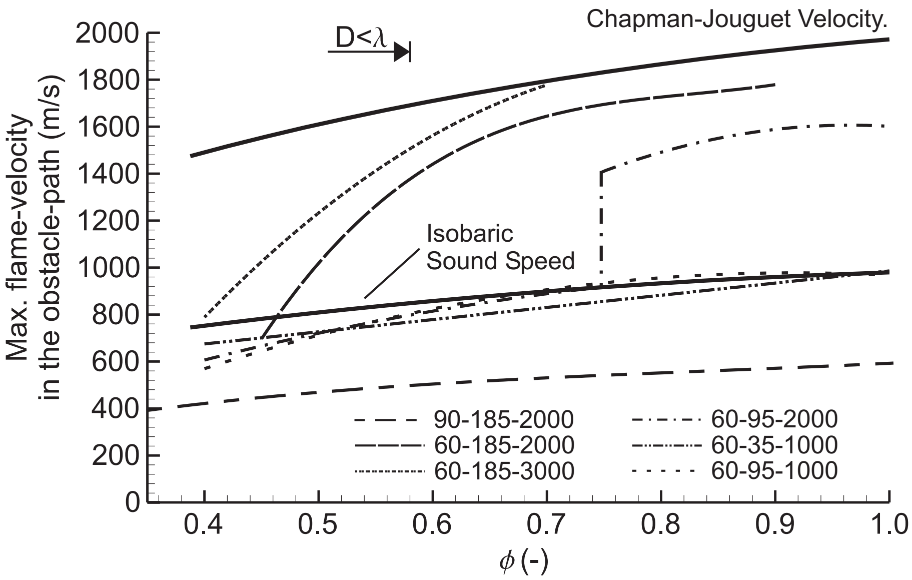

Figure 1: Maximum flame-velocity in the obstacle-path for six obstacle configurations. Notation for the obstacle is: blockage-ratio BR – spacing l_{SP} – length of obstacle-path L_{OP}.

occurrence of various combustion modes in tubes: The maximum deflagrative flame velocity in these experiments was limited by the isobaric sound speed and quasidetonation occurred only, if the diameter of the unblocked obstacle section was larger than the detonation cell-width. These criteria were achieved with an obstacle spacing commensurate to the tube-diameter. These criteria could only be verified for similar geometrical conditions. If a larger obstacle spacing is used (e.g. configuration “60-185-2000” in Fig. 1), flame-velocities in the range between the isobaric sound speed and the Chapman-Jouguet (CJ) are observed for mixtures, considerably less sensitive as required for the onset of a quasidetonation according to the criteria of Peraldi et al. This result leads to the assumption that the propagation of fast deflagrations and detonations depends very strongly on the obstacle configuration within the tube. One explanation of these high flame-velocities is that the momentum losses become smaller due to the larger distance of the obstacles. On the other hand, the obstacles together with the relatively small dimension of the tube maintain a system of strong transverse shock-waves, which serve as permanent hot (ignition) spots behind the leading, longitudinal shock wave. Therefore, this flame-propagation mode can be interpreted as a superposition of a fast deflagrative and a shock-induced quasi-detonative one.

Flame propagation in the unblocked tube

In Figure 2, the dependence of the combustion mode at the beginning of the unblocked tube section on the equivalence ratio ϕ as well as the obstacle configuration is shown. The main point of interest was, under which conditions a marginal detonation could be observed. A comparison of Fig. 1 and 2 shows that the onset of a detonation at the $D = \lambda/\pi$ detonation-limit was only observed for obstacle configurations in which higher velocities than the isobaric sound speed were observed at the respective mixture composition. Whenever only a maximum flame-velocity up to the isobaric sound speed was achieved within the obstacle path, the transition from deflagration to detonation occurred exclusively at the $D = \lambda$ limit for the onset of a planar detonation front.

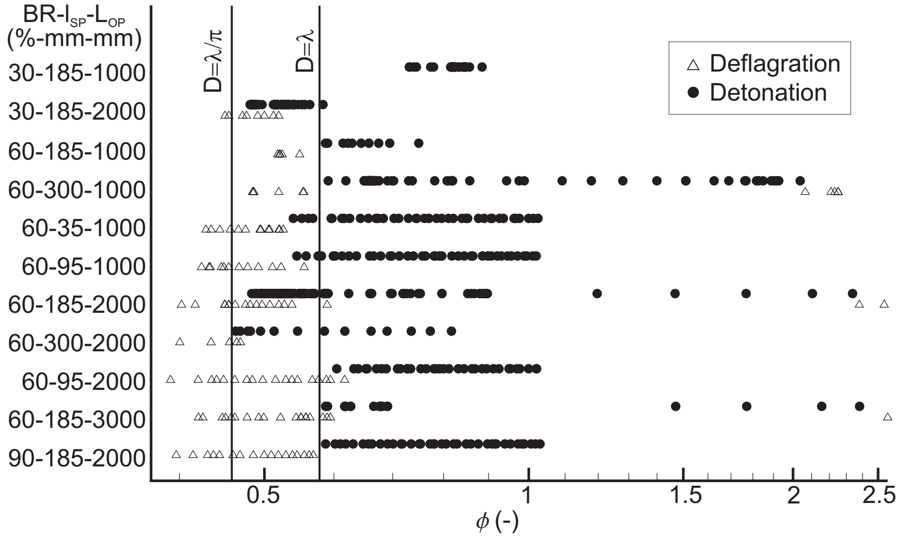

Figure 2: Detonation onset right after the obstacle path for investigated obstacle configurations.

For mixtures between the limits $\lambda/\pi < D < \lambda$ it was observed that three different combustion modes coexist after the flame has left the obstacle-path for configurations with a large obstacle spacing: One possibility is, that the flame-front decouples from the leading shock wave and propagates further as a slow deflagration. On the other hand, it is possible that the flame front remains coupled to the shock-front and transits into a CJ-Detonation. Furthermore, it was observed that the shock/flame-system remains coupled an propagates as a fast deflagration with a constant velocity of approx. 1000 m/s up to the end of the tube. All these transition processes have been investigated by means of the color-schlieren technique (results can be downloaded from the URL given at [Ede01] and will be presented in the full paper).

In order get a statistical figure about the occurrence of each combustion mode within this mixture range, over 250 experiments have been conducted for the configuration "60-185-2000". The probability-distribution is shown in Fig. 3.

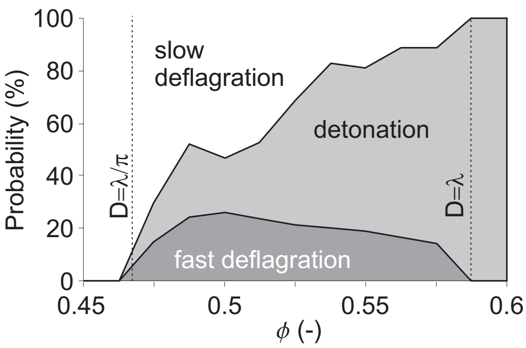

Figure 3: Probability for detonation-onset within the mixture range corresponding to $\lambda/\pi < D < \lambda$.

The probability for the onset of a fast deflagration in this mixture range is up to 20% and above the $D = \lambda$ limit, only the transition to the detonative combustion mode occurred. These fast propagating deflagrations are of high importance, as they generate the highest pressure loading (more than 200 bar have been observed) if reflected at a wall, cf. [EGM99]. The planar laser-induced predissociation fluorescence has been applied to identify theses various propagation modes. Figure 4 shows the OH-radical distribution of a fast deflagration and two detonations within the mixture range corresponding to $\lambda/\pi < D < \lambda$. For the fast-deflagration case, a clearly defined flame-front as well as a simultaneous reaction zone without a defined flame-contour can be identified, Fig. 4-A. The reaction-zone-structure indicates therefore that this propagation mode has to be close to regime of the

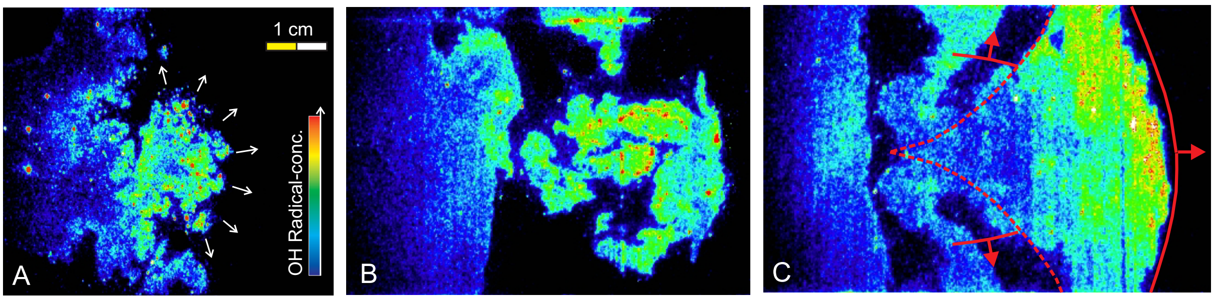

Figure 4: OH-radical distribution of a fast deflagration (A, $\phi = 0.51$), and two marginals detonations (B: $\phi = 0.49$; C: $\phi = 0.57$).

well-stirred reactor according to the phase diagram of Borghi and Peters [Pet97], which could also be shown by analytical considerations. In case of detonations within this mixture-range, it could be observed that large pockets of unreacted gas are formed behind the shock-induced reaction zone, which has also been shown by numerical studies of Oran [Ora99]. If the tube-diameter is smaller than the detonation cell-width, no cellular detonation front structure was detected. By means of self-fluorescence measurements it could be shown, that only one transverse wave exists. Due to the large reaction-zone length, the ignition is not only triggered by the ignition-delay behind the shock-waves. Furthermore, fluctuations become more and more dominant which lead in the case of hydrogen-air mixtures to such an undefined reaction-zone structure as shown in Fig. 4-B. If the mixture is more sensitive so that the detonation-cellwidth is approximately equal to the tube-diameter, it was observed that one detonation cell fits perfectly into the tube, Fig. 4-C. Nevertheless, due to the high ignition delay, the ignition behind the mach-stem occurs much earlier than behind the normal shock wave and because of the strong curvature of the shock-system, unreacted pockets are formed, too.
The transition-process to the CJ-detonation mode was not only observed directly behind the obstacle-path. Furthermore it was observed that detonation onset occurred after the flame front has propagated over a length of some meters as a fast deflagration. The necessary condition for this incident was that the $D = \lambda$ criterion was met. The detonation-onset occurred in this case

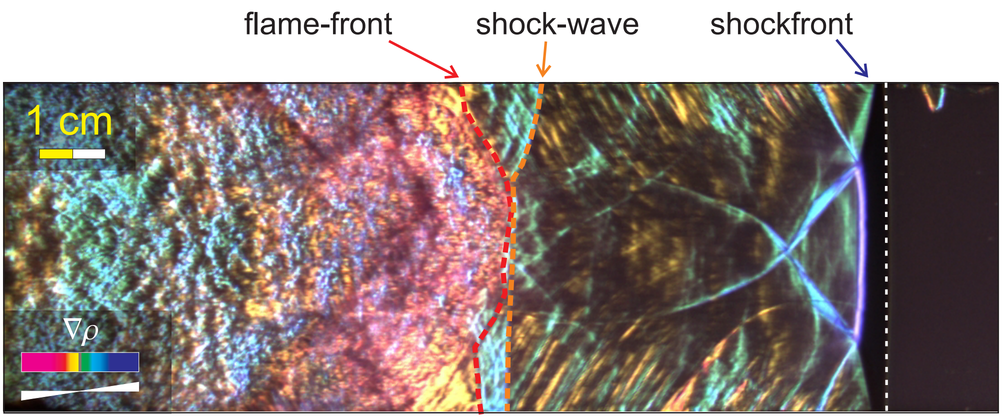

Figure 5: Detonation onset within the turbulent flame brush. Hydrogen-air mixture ($\phi = 0.65$), flame propagation velocity: 1050 m/s.

only at the flame front in a way that a shock-wave was generated in the flame-brush and the flame itself immediately couples to this shock-wave as shown in Fig. 5.
Even in an entirely empty tube, a transition from deflagration to detonation was observed. The transition could be attributed to a spontaneous flame-acceleration induced by a Richtmyer-Meshkov instability that accelerated the flame up to the isobaric sound speed. In the moment, when the shock-wave passed the flame-front, the highest reaction rates (even higher than for detonations) were detected. After the flame acceleration, induced by the Richtmyer-Meshkov instability, a detonation onset within the flame-brush occurred as shown in Fig. 5.

Influence of additives

Besides pure hydrogen-air mixtures, the influence of steam, carbondioxid, carbonmonoxide, and methane was observed. The influence of steam and carbondioxid was quite similar and confirmed the inhibitoric influence of these additives. In both cases, no detonation occured when adding more than 15 vol % of steam or carbondioxide respectively. The additive carbonmonoxide in the hydrogen-air-carbonmonoxide-mixture showed the same combustion-behavior as hydrogen. Therefore, if the entire equivalence-ratio is concerned, the mole-fraction of carbonmonoxid can be replaced by hydrogen for safety analysis reasons.
The most unexpected combustion-behavior was observed for the so-called hythane (hydrogen-methane-air) mixtures. Although the detonation-cellwidth of pure methane-air mixtures is known to be several times larger than that for hydrogen-air mixtures, a minimum fuel-concentration for the detonation onset was observed for hythane mixtures. For these experiments, the equivalence ratio ϕ was held constant and the hydrogen-methane molefraction was varied. For $\phi = 1.0$ and $\phi = 0.8$, no difference in the combustion behavior was detected. Due to the lower equivalence ratio of methane-air mixtures, the minimum fuel molefraction required for a detonation onset is 4 vol % lower for a $\phi = 0.8$ hythan-mixture than the minimum required mole-fraction for pure hydrogen-air mixtures. Therefore, hythane-mixtures are more dangerous than pure hydrogen-air mixtures, if the detonability of both mixtures is concerned.

References

[BCD+00] W. Breitung, C.K. Chan, S. Dorofeev, A. Eder, B. Gerlfand, M. Heitsch, R. Klein, A. Malliakos, J.E. Shepherd, E. Studer, and P. Thibault. Flame Acceleration and Deflagration-to-Detonation Transition in Nuclear Safety. OECD State-of-the-Art Report by a Group of Experts, NEA/CSNI/R(2000)7, 2000.

[Ede01] A. Eder. `http://www.td.mw.tum.de/~eder/colorschlieren.html`, 2001.

[EGM99] A. Eder, C. Gerlach, and F. Mayinger. Determination of quantitative criteria for the transition from deflagration to detonation in H_2/air-mixtures. In *Proceedings of the 22nd Int. Symp. on Shock Waves (ISSW22)*. London, 1999. ISBN 085432-706-1.

[Ora99] E.S. Oran. The structure of propagating detonations: Lessons from numerical simulations. In *Proc. of the 17th Int. Coll. on the Dynamics of Explosions and Reactiv Systems (ICDERS)*. Heidelberg, 1999. ISBN 3-932217-01-2.

[Pet97] N. Peters. Four lectures on turbulent combustion. In *ERCOFTAC Summer School*, Aachen, Germany, September 1997.

[PKL86] O. Peraldi, R. Kystautas, and J.H.S. Lee. Criteria for transition to detonation in tubes. In *21 st (Int.) Symposium on Combust.*, pages 1629–1637. The Combustion Institute, 1986.

[UO66] P.A. Urtiew and A.K. Oppenheim. Experimental observations of the transition to detonation in an explosive gas. In *Proc. Roy. Soc. London*, volume A295, pages 13–28. The Royal Society, 1966.