Deflagration-to-Detonation Transition in Highly Reactive Combustible Mixtures

M. A. Liberman[1, 2], M. F. Ivanov[3], A. D. Kiverin[3], M. S. Kuznetsov[4], A. A. Chukalovsky[2], T. V. Rakhimova[2]

[1]Skobeltsyn Institute of Nuclear Physics, Moscow State University, Moscow, 119991 Russia
[2] Department of Physics, Uppsala University, Box 530, 751 21, Uppsala, Sweden
[3] Joint Institute for High Temperatures, Russian Academy of Science, Moscow, Russia
[4]Institute for Nuclear and Energy Technologies, Karlsruhe Institute of Technology, Kaiserstarsse, 12, 7613, Karlsruhe, Germany

1 Introduction

The mechanism by which a deflagration transforms into a detonation remains one of the most interesting unresolved problems in combustion theory. After work [1] where Zel'dovich et al. in 1970 have shown that the nonuniform temperature distributions may initiate a detonation, it was suggested that the mechanism of the transition from deflagration to detonation (DDT) is a reactivity gradient (usually the temperature gradient) which is involved in the Zel'dovich criterion of detonation initiation. This seemed quite plausible and in the 40 years since that time many researchers have done an enormous amount of computations in attempt to explain DDT using the idea of the gradient mechanism. However the very fact is that capability of the temperature gradient to initiate detonation was demonstrated only for the one-dimensional models with a prescribed nonuniform temperature distribution. More important is that the gradient concept and the multidimensional numerical studies of DDT were heavily based on a simplified chemical model in which the reaction proceeds via a one-step Arrhenius kinetics [2]. Recently Sharpe and Short [3] used a two-step kinetics model which mimics chain-branching chemistry and found that this leads to a profound effect on the criteria of a temperature gradient required for the detonation initiation, which a one-step reaction model cannot reproduce. Comprehensive studies of the evolution to detonation from the temperature nonuniformities with a detailed chemical kinetics [4] show that the evolution to detonation is profoundly different for detailed chain branching kinetic models than for one-step kinetic models and it is different in different fuels such as hydrogen-air or methane-air. The steepest temperature gradient capable to initiate detonation is by orders of magnitude shallower compared to that predicted from a one-step model. It is therefore important to investigate the differences in the DDT process between chain-branching kinetics and the predictions from simple one-step models. In the present study we present numerical simulation of DDT in highly reactive hydrogen-oxygen mixtures using a detailed chemical reaction mechanism. We show that origin of the transition from deflagration to detonation is not the gradient mechanism and the new proposed mechanism of DDT is

Correspondence to: M.A. Liberman <misha.liberman@gmail.com>

considerably different. Results of the numerical simulations are consistent with extensive experimental studies of DDT in hydrogen-oxygen and ethylene-oxygen mixtures.

2 Problem formulation and numerical results

The high resolution simulations modeled a flame ignited at the closed end and then propagating to the open end of the two-dimensional rectangular channel with smooth no-slip walls. The computations solved the two-dimensional, time-dependent, reactive Navier-Stokes equations for compressible flow including the effects of viscosity, thermal conduction, molecular diffusion, and detailed chemical kinetics for the reactive species H_2, O_2, H, O, OH, H_2O, H_2O_2, and HO_2 with subsequent chain branching, production of radicals and energy release [5, 6]. The real equations of state for the fresh mixture, the reactive species and combustion products were taken with the temperature dependence of the specific heats, heat capacities and enthalpies of each species borrowed from the JANAF tables. The viscosity and thermal conductivity coefficients of the mixture were calculated using the Lennard-Jones potential. Coefficients of the heat conduction of i-th species $\kappa_i = \mu_i c_{pi} / \mathrm{Pr}$ are expressed via the kinematic viscosity μ_i and the Prandtl number $\mathrm{Pr} = 0.75$. The reaction scheme for a stoichiometric H_2-O_2 mixture used in the simulations has been tested in many applications and proved to be adequate to complete chemical kinetic scheme. The computed thermodynamic, chemical, and material parameters using this chemical scheme are in a good agreement with the flame, shocks and detonation wave characteristics measured experimentally. The computational method was described and tested extensively in [5, 6]. Its validation, convergence and resolution tests taken to resolve the structure of the flame front with the meshes up to 64 computational cells per flame width are presented in [7].

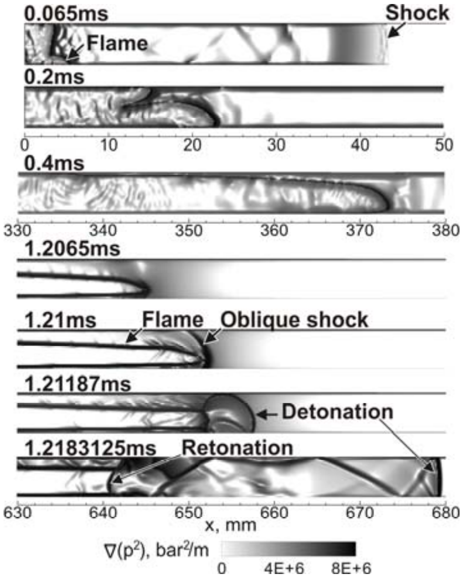

Figure 1: Temporal evolution of the reaction wave during transition to detonation in channel of width D=5mm for hydrogen oxygen, $P_0 = 1\mathrm{bar}$. Detonation starts after 1.21ms.

Figure 1 shows evolution of the overall flow and flame front development during transition to detonation in the channel of width D=5mm with H_2-O_2 mixture at initial pressure $P_0 = 1\mathrm{bar}$; the computational time instants are shown on the left side of each frame. Shortly after the ignition, but when the details of the initiation are already `forgotten', the flame develops either a tulip shape or a bulge extended along the channel walls if at the beginning small perturbations were imposed. At the beginning the accelerating flame produces shocks far ahead from the flame front. Thereupon the shocks are produced in the vicinity adjacent to the flame (from 0.5ms). The further evolution leads to a localized autoignition and triggering detonation in about 1μs after t=1.21ms. The last frame in figure 1 shows the detonation wave with cellular structure and retonation wave at the left of the computational domain.

A principal feature of the flame acceleration in tubes with no-slip walls which determines the mechanism of DDT is the formation of a compressed pocket of unreacted gas adjacent ahead to the flame. A high density unreacted mixture of a compressed pocket enters in the high temperature reaction zone of the flame producing the large amplitude pressure pulse at the flame front. The increased pressure in the reaction zone increases rate of the reaction. As a result of the positive feedback between the increasing pressure pulse and the reaction rate, the pressure peak at the flame front grows exponentially in time, steepens into a strong shock which is coupled with the reaction zone forming the overdriven detonation wave.

Figure 2 (left) shows evolution of the temperature profile in the flame and the pressure peak formation for the conditions of figure 1. It should be remarked that temperature in the compressed pocket of unreacted gas ahead to the flame (preheat zone) does not exceed 550K, which means that reaction in the preheat zone is totally frozen. Figure 2 (right) shows the computed evolution of the pressure peak, where the dashed line corresponds to the approximation $P_{max} \propto P_0 \exp(1.54 \cdot t / ms)$. By the time 1.21ms the amplitude of the pressure peak becomes large enough and the pressure pulse steepens explosively into the strong shock. After this moment the transition to detonation occurs in less than 1μs. It should be noted that the pressure peak at the flame front reaches 14bar at the time 1.2ms and it increases above 20bar at the time 1.21ms when the transition to detonation starts. Such amplitude of the pressure peak is comparable to a pressure jump across the leading shock in a detonation wave.

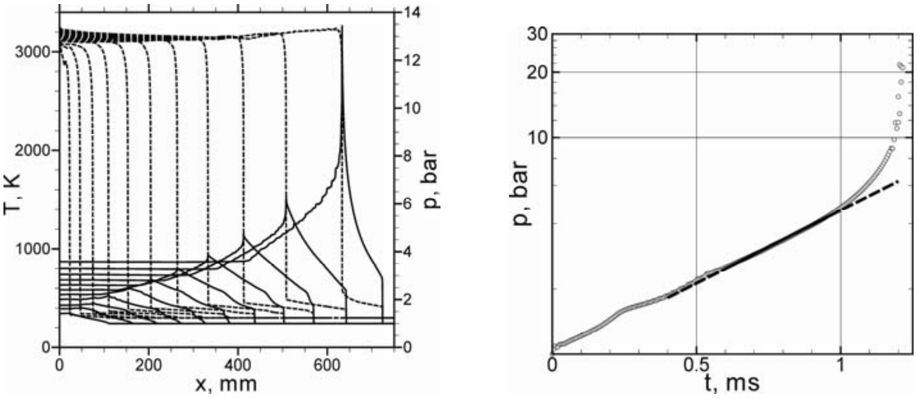

Figure 2 (left): Sequence of temperature (dashed line) and pressure (solid line) profiles corresponding to leading point in the flame front represents the flame structure and the pressure peak formation for the conditions of figure 1. Times are from 0.2ms to 1.2ms with interval 0.1ms. (Right): computed maximum of the pressure pulse. Dashed line corresponds to $P_{max} \propto P_0 \exp(1.54 \cdot t / ms)$.

The last stage of the actual transition to detonation is shown in figure 3, which presents variations of the pressure temperature and concentration Y_H of H-radicals profiles at sequential times, from t=1.2101875ms till 1.2113125ms with the time interval 0.125μs. The transition to detonation is clearly seen from the increased temperature of the products and the temperature gradient. One can see that the distribution of H-radicals in the reactions is different during the earlier times of a deflagration where the H-radicals appears within the front of the deflagration wave and after the transition to detonation, which consists of the well pronounced shock wave with the jump in temperature and pressure followed by the reaction zone.

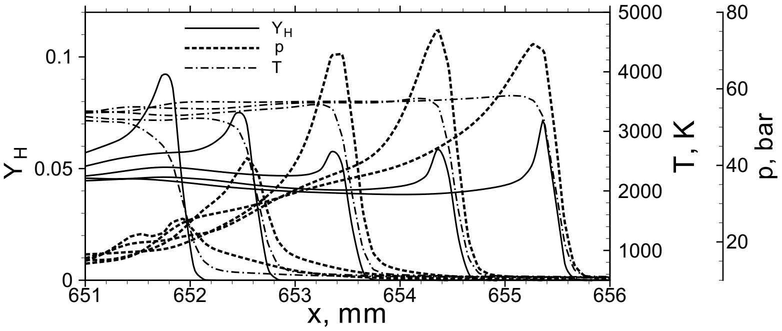

Figure 3: Sequence of temperature and pressure profiles during the transition to detonation in H_2-O_2 from 1.2101875ms till 1.2113125ms; time step is 0.125μs.

3 Experiments

The experimental studies of DDT in a highly reactive stoichiometric hydrogen-oxygen and ethylene-oxygen mixtures are focused on the key characteristics of DDT: 1) the features of the flame acceleration which define strength and location of the shocks generated by accelerating flame, 2) the formation of a pocket of compressed unreacted mixture (the preheat zone) adjacent to the flame during the next stage of the flame acceleration.

The experimental facility and the optical Schlieren system with stroboscopic pulse generator and high-speed camera used to record the combustion wave evolution were described in [8] and the references within. A sequence of shadow photos in figure 4 taken with interval 0.1ms shows the evolution of the flow during the second stage and the final stage of transition to detonation in ethylene-oxygen mixture, $P_0 = 0.12\text{bar}$. In contrast to the earlier stages, the compression waves produced by the accelerating flame steepen into shocks, forming a compressed pocket of unreacted material between the shock and the flame. In the second image, at 3.85ms, the shocks are coalesced creating a pocket of compressed unreacted gas which appears adjacent ahead to the flame. On the last frame one can see already formed detonation and retonation waves.

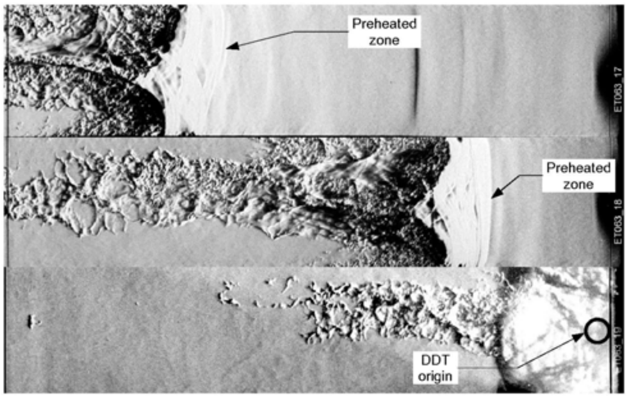

Figure 4: Sequence of shadow photographs show during the second stage and transition to detonation in ethylene-oxygen mixture, $P_0 = 0.12\text{bar}$. The frames are at: 3.75; 3.85; 3.95ms from the top.

4 Conclusions

The present study reveals a principle feature of flame acceleration in tubes with no-slip walls leading to the formation of the pocket of compressed unburned material adjacent ahead to the flame front, which is the key phenomenon for the transition to detonation. The pocket of compressed unburned material adjacent ahead to the flame results in the birth and growth of a large amplitude pressure pulse which grows exponentially and steepens into a strong shock which is capable to trigger detonation. This mechanism of a detonation initiation has been overlooked previously mainly because of a one-step reaction model cannot correctly reproduce the main properties of the combustion and the induction time in chain-branching kinetics. The previous attempts to explain the mechanism of DDT by the gradient mechanism have used simplified one-step chemistry with low activation energy which at least for highly reactive mixtures is not small in the range below 1000 K. This might mask the actual mechanism of DDT sufficiently for it to not manifest itself. It is shown that a detailed chemical model has a profound effect on the validity of the DDT through gradients of temperature.

The proposed new mechanism of DDT is the formation of a large amplitude pressure pulse in the front edge of the flame. The pressure is amplified due to a positive feedback between the pressure rise and the enhanced reaction, growing exponentially and steepens into a strong shock which is coupled with the reaction zone and forms overdriven detonation wave. It must be stressed that the obtained results arise from extensive highly resolved numerical simulations using a detailed chemical reaction mechanism. The validity of the described mechanism of DDT is confirmed by the experimental studies. The obtained results make questionable applicability of the gradient mechanism as a mechanism of the transition from deflagration to detonation. In real combustible systems, the temperature gradients, which appear in the form of hot spots and the like, do not satisfy the criteria necessary to initiate detonation. As such, the results should set the groundwork for future computations. Similar simulations for slow reactive mixtures, e.g. methane-air are in progress and will be reported elsewhere.

References

[1] Zeldovich, Ya.B. et al. (1970) On the Development of Detonation in a Non-Uniformly Preheated Gas Astronaut. Acta, Vol. 15:313-324.

[2] Oran, E. S., Gamezo V. N. (2007) Origins of the deflagration-to-detonation transition in gas-phase combustion, Combust. Flame, Vol. 148: 4-47.

[3] Sharpe, GJ and Short, M. (2003) Detonation ignition from a temperature gradient for a two-step chain-branching kinetics model, J. Fluid Mech. 476: 267-292.

[4] Kiverin, A.D., Chukalovsky, A.A., Ivanov, M.F., Liberman, M.A., *Detonation Initiation by a Temperature Gradient for a Detailed Chemical Reaction Models*, 2011 23rd ICDERS, submitted.

[5] Liberman, M.A. et al. (2010) Deflagration-to-detonation transition in highly reactive combustible mixtures, Acta Astronautica, Vol. 67: 688-701.

[6] Liberman, M.A. et al. (2010) On the Mechanism of the Deflagration-to-Detonation Transition in a Hydrogen–Oxygen Mixture, Zh. Exp. Theor. Fiz. (JETP) 138: 772-788.

[7] Ivanov, M.F., A. D. Kiverin A.D., Liberman, M.A. (2010) Flame Acceleration and Deflagration-to-Detonation Transition in Stoichiometric Hydrogen/Oxygen in Tubes of Different Diameters, Combust. Flame, submitted.

[8] Kuznetsov, M., Liberman, M., Matsukov I. (2010) Experimental Study of the Preheat Zone Formation and Deflagration to Detonation Transition, Comust. Sci. Tech. 182: 1628-1644.