DEFLAGRATION-TO-DETONATION TRANSITION IN HYDROGEN OXYGEN MIXTURE WITH A DETAILED CHEMICAL REACTION MECHANISM

M. A. Liberman[1, 2*], **M. F. Ivanov**[3], **A. D. Kiverin**[3]
[1] **Department of Physics, Uppsala University, Box 530, 751 21, Uppsala, Sweden,**
misha.liberman@gmail.com
[2] **Skobeltsyn Institute of Nuclear Physics, Moscow State University, Moscow, 119991 Russia,**
mliberman@mics.msu.su
[3] **Joint Institute for High Temperatures, Russian Academy of Science, Moscow, Russia,**
ivanov_mf@mail.ru (Ivanov), alexeykiverin@gmail.com (Kiverin)

ABSTRACT

High resolution numerical simulations used to study the mechanism of deflagration-to-detonation transition (DDT). The computations solved two-dimensional, time-dependent, reactive Navier-Stokes equations including the effects of compressibility, molecular diffusion, thermal conduction, viscosity and detailed chemical kinetics for the reactive species with subsequent chain branching, production of radicals and energy release. It is shown that from the beginning the flame accelerates exponentially producing shock waves far ahead. On the next stage the flame acceleration decreases and the shocks are formed close ahead of the flame front. The final stage is the actual transition to detonation. During the second stage a compressed unreacted mixture of increased density enters the flame producing a high pressure pulse which enhances reaction rate and the heat release in the reaction zone with a positive feedback coupling between the pressure pulse and the reaction rate. As a result the peak of the pressure pulse grows exponentially, steepens into a strong shock which is coupled with the reaction zone forming the overdriven detonation. This new mechanism of DDT is different from the Zel'dovich's gradient mechanism. The temperature gradients, which appear in the form of hot spots and the like, are not suitable to initiate detonation.

1. INTRODUCTION

The mechanism by which a deflagration transforms into a detonation remains one of the most interesting unresolved problems in combustion theory. In 1970 Zel'dovich et al. [1] have shown that the prescribed nonuniform temperature distributions may initiate a detonation. In the 40 years since that time many workers have done an enormous amount of computations in attempt to explain transition from deflagration-to-detonation (DDT) using the idea of the Zel'dovich's gradient of detonation initiation. Though this idea seems quite natural as a mechanism of DDT, the very fact is that capability of the temperature gradient to initiate detonation was demonstrated only in the one-dimensional models with a prescribed nonuniform temperature distribution. More important is that the Zel'dovich's 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]. It is well known that a one-step reaction model cannot reproduce the main properties of the combustion such as the induction time in chain-branching kinetics. Recently Sharpe and Short [3] used a two-step kinetics model, which to some extend mimics features of chain-branching chemistry. They found that this leads to a profound effect on the criteria of a temperature gradient required for the detonation initiation. Comprehensive studies of the evolution to detonation from the temperature nonuniformitiesis 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 that it is different in different fuels, such as hydrogen-air or methane-air. The steepest temperature gradient capable to initiate detonation is by a few orders of magnitude shallower compared to what predicted from a one-step model. This means that a temperature gradient which was suggested to appear in the form of hot spots or the like should be much too long and very unlikely to be origin of the DDT. We proposed new mechanism of DDT which is considerably different form the gradient mechanism. It is shown that the new mechanism is consistent with experimental studies of DDT in hydrogen-oxygen and ethylene-oxygen mixtures.

2. FORMULATION AND RESULTS OF SIMULATIONS

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 including the effects of compressibility, convection, 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 reactive species and combustion products were taken with the temperature dependence of the specific heats 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 with 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 kinetics scheme. The computed thermodynamic, chemical, and material parameters using this chemical scheme are in a good agreement with the flame and detonation wave characteristics measured experimentally. The computational method was described and tested extensively in [5, 6]. The convergence and resolution used the meshes up to 64 computational cells per flame width to resolve the structure of the flame front.

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 times 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 close vicinity of 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.

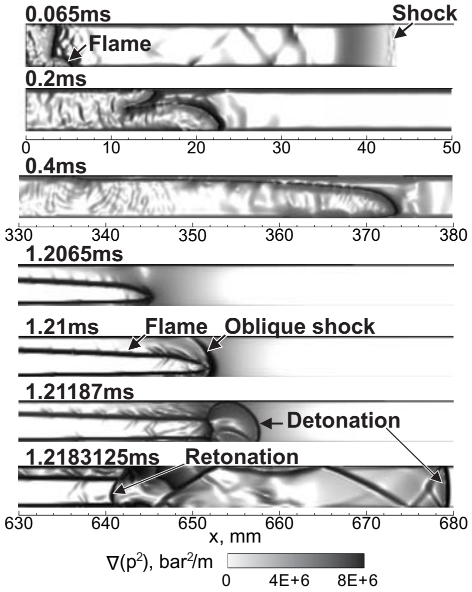

Figure 1. Temporal evolution of the reaction wave during transition to detonation in channel D=5mm for hydrogen oxygen, $P_0 = 1\text{bar}$. Detonation starts after 1.21ms.

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 pressure pulse of large amplitude 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 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. This mechanism of DDT is considerably different from the gradient mechanism of DDT.

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 noted that temperature in the compressed pocket of unreacted gas adjacent 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 is $P_{\max} \propto P_0 \exp(1.54 \cdot t / \text{ms})$. By the time 1.21ms the amplitude of the pressure peak becomes large enough, and the pressure pulse steepens explosively into the strong shock and after this moment the transition to detonation occurs in less than 1μs. It should be remarked 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 shock with the Mach number 5, which is close to the strength of the leading shock in a detonation wave. 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.

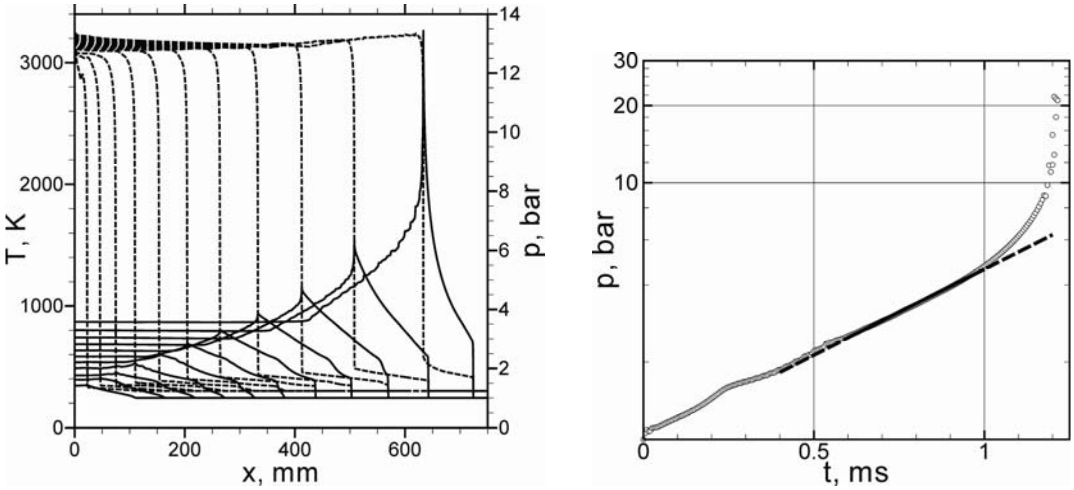

Figure 2 (left): Sequence of temperature (dashed line) and pressure (solid line) profiles corresponding to leading point in the flame front represents the flame structure and the pressure peak formation for the conditions of figure 1. Times are from 0.2ms to 1.2ms with interval 0.1ms. (Right): computed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

One can see that the distribution of H-radicals in the reactions is different during the earlier times of a deflagration regime and after the transition to detonation. In deflagration the H-radicals appear within the front of the deflagration wave, while structure of the detonation wave consists of the well pronounced shock wave with the jump in temperature and pressure following by the reaction.

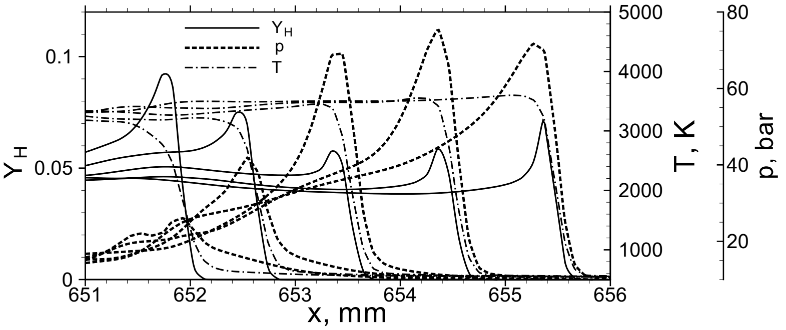

Figure 3: Sequence of temperature and pressure profiles during the transition to detonation in H_2-O_2 from 1.2101875ms till 1.2113125ms; time step is 0.125μs.

3. EXPERIMENTAL STUDIES

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 adjacent to the flame, the preheat zone, 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 [7] and references within. A sequence of shadow photos in figure 4 taken with interval 0.1ms shows the evolution of the flow during the last stage and the transition to detonation for the initial pressure $P_0 = 0.12\text{bar}$. In contrast to the earlier stages, the flame generates the compression waves which steepen to shocks almost at the flame front. The shocks ahead of the flame are formed very close to the flame front, producing 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 close ahead to the flame. On the last frame we see already detonation and retonation waves.

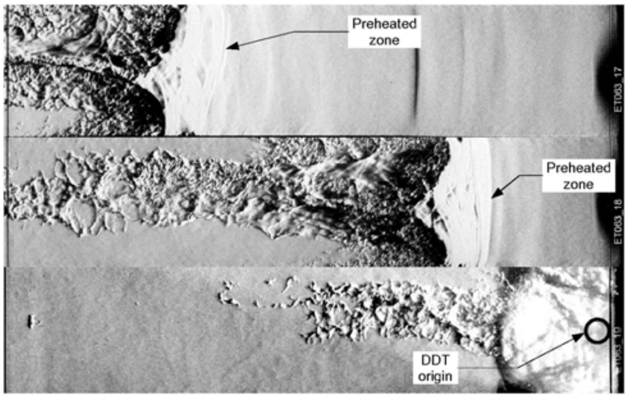

Figure 4: Sequence of shadow photographs show during the second stage and transition to detonation in ethylene-oxygen mixture, $P_0 = 0.12\text{bar}$. The frames are at: 3.75; 3.85; 3.95ms from the top.

4. CONCLUDING REMARKS

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 deflagration-to-detonation transition. The pocket of compressed unburned material adjacent ahead to the flame results in the birth and growth of a large amplitude propagating 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 properties of the combustion and the induction time in chain-branching kinetics. The previous attempts to explain origin of DDT by the detonation initiation through the gradient mechanism [2] have used a one-step chemistry with low activation energy which might mask the actual mechanism of DDT sufficiently for it to not manifest itself. 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. The pressure peak grows 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 the highly resolved numerical simulations using a detailed chemical reaction mechanism. The validity of the described mechanism of DDT is confirmed by the extensive experimental studies of DDT in highly reactive mixtures. As such, the results should set the groundwork for future computations. The obtained result makes questionable applicability of the gradient mechanism as origin 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, are too steep, if any, to initiate detonation. It was shown in [8] that taking into account a detailed chemical model, the minimal length of the steepest temperature gradient capable to initiate detonation is by many orders of magnitude shallower than that predicted within a frame of a single-step chemical model.

REFERENCES.

[1] Ya.B. Zeldovich, et al., *Astronaut. Acta*, 15 (1970) 313-324.

[2] E. S. Oran, V. N. Gamezo, *Combust. Flame*, 148 (2007) 4-47.

[3] G.J Sharpe and M. Short, *J. Fluid Mech*. 476 (2003) 267-292.

[4] M. A. Liberman, A. D. Kiverin, A. A. Chukalovsky, M. F. Ivanov, *Detonation Initiation by a Temperature Gradient for a Detailed Chemical Reaction Models*, 2011 7th US National Combustion Meeting.

[5] M. A. Liberman, et al., *Acta Astronautica*, 67 (2010) 688-701.

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

[8] M. A. Liberman, A. D. Kiverin, M. F. Ivanov (2011) On Detonation Initiation by a Temperature Gradient for a Detailed Chemical Reaction Models, *Phys, Letters* A 375, 1803-1808.