On the structure of combustion waves in poorly premixed gaseous mixture

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Abstract.
The paper studies numerically the features of combustion wave propagation in poorly premixed hydrogen-oxygen mixture. In present study poorly premixed mixtures are taken to be those filling the combustor volume with random deviations of components concentrations from the average values on the spatial scales of the order of 0.1 – 1.0 mm. The numerical analysis shows that at relatively small spatial scales of concentration non-uniformities (below 0.5 mm) and at relatively low dispersion of concentration the rate of flame acceleration increases that leads to faster transition to detonation compared with the case of ideally premixed mixture. Enhancement of flame acceleration is related with continuous formation of perturbations of the flame front, among which the greatest impact belongs to those with short wavelengths. On the one hand these perturbations intensify the evolution of Darrieus-Landau instability and on the other hand act on the flame front in similar way as the turbulent pulsations do. At larger spatial scales of the non-uniformities or at increased dispersion of concentrations the flame acceleration rate decreases since the greater role in the process evolution now belongs to chemical factor associated with concentration deviation from the stoichiometric compound.

1. Introduction
One of the most hazardous processes arising during the severe accidents on the nuclear power plants or other industrial sectors involving hydrogen technologies is the explosion of hydrogen accidentally released into the atmosphere. Ejected hydrogen fills the technical rooms and stirs with air forming a flammable mixture. Due to the paramount importance for explosive safety the issue of hydrogen release into the air and its subsequent combustion is widely studied in number of works. As a rule these works consider jets of pressurized hydrogen in the air environment and ignition of resultant so-called non-premixed mixtures. In this case the dynamics of hydrogen jet is determined by the pressure inside the hydrogen-containing vessel and by the size of outlet orifice through which hydrogen flows into the air [1, 2, 3]. Volumetric hydrogen release into the atmosphere is less common but still possible situation. When large amounts of hydrogen releases at relatively low overpressure one can expect formation of volumetric clouds of stirring air and hydrogen. After the release mixture components spatial distributions evolve through the several stages from the fast turbulent mixing to the slow diffusive relaxation. On the final stage the large-scale structures containing different components have already dissipated however the mixture is still locally non-uniform. The relaxation to the fully uniform state could last for a relatively long time compared with characteristic times of ignition and flame propagation through
the mixture. For example, in [4] nearly uniform mixture inside spherical vessel of 2 m radii was formed only after relaxation of convective and diffusive processes lasting for 30 minutes after turbulent stirring of separately fed hydrogen and air. Even longer life time of small concentration non-uniformities could be observed during accidental accumulation of combustible mixture in volumes (e.g., accumulation of hydrogen-air mixture in the upper part of the reactor room at the nuclear power plant). The origin of such accumulation is related with slow leakage of one of the components and its subsequent relatively slow redistribution in the volume due to convection and diffusion. The combustion initiated at the stage of mixture composition relaxation would propagate like in premixed case but under the action of certain disturbances due to the non-uniform distribution of fuel concentration. Further let call such a situation as combustion of poorly premixed mixture.

Although the applied significance of poorly premixed mixtures combustion is evident it is still less studied compared with combustion of fully premixed mixtures. The main problem is the turbulent nature of the stirring process in most of practically important cases. This determines a high complexity and unpredictability of local spatio-temporal evolution of the concentration field structure defined by convection, viscous dissipation and diffusive smoothing of concentration fluctuations. Nowadays most of works devoted to non-uniform mixtures combustion are mainly associated with initial stage of combustible mixture formation. As a rule both experimental and theoretical research works in this field are carried out in the simplified problem setup where the fuel concentration is distributed via gradient, orthogonal [5, 6] or oriented along the direction of flame propagation [7, 8, 9, 10]. However even the results obtained in such simple cases allow to elucidate the features intrinsic to the process of flame propagation under the conditions of fuel stratification. In this paper let consider numerically the features of flame propagation in poorly premixed mixture which state can be treated as one corresponding to the stage of non-linear stabilization and decay of non-uniformities formed in the process of stirring. By this moment a wide spectrum of concentration perturbations is formed with wavelengths from mixing zone width down to tenths of millimeter. The leading role in the process of stirring and formation of these perturbations in mixture components concentrations belongs to the joint action of Rayleigh-Taylor and Kelvin-Helmholtz instabilities. Mentioned estimation of the range of wavelengths agrees well with experimental data [11]. Obviously, on the stabilization stage the basic mechanism of concentration perturbations decay is the diffusion while the main role in perturbations support belongs to the residual inertia of convective flows and the slow leakages of the mixture components. Taking into account that the stirring represents a turbulent process and therefore on the final stages of stirring the mixture components are distributed chaotically it is reasonable to consider combustion in the mixture with initially given concentrations fields characterized by random deviations of the fuel and oxidizer concentrations from the averaged values.

2. Problem setup and numerical algorithm
To estimate the degree of influence of concentrations perturbations on the processes involved in gaseous combustion let consider as the example the problem of flame propagation through two-dimensional channel filled with stoichiometric hydrogen-oxygen mixture. Chosen problem setup is conventional and close to that considered numerically in [12]. The solution provides all the stages of flame acceleration including transition to detonation that allowed us to analyze in details the basic mechanisms determining both flame acceleration and transition to detonation [12]. Model of chemical kinetics used here is not the same as in [12] and spatio-temporal scales of deflagration-to-detonation transition (DDT) are slightly different but the qualitative features of flame evolution remained unchanged.

Gasdynamics of the gaseous mixture combustion was modeled via solving governing equations system based on Navier-Stokes equations for multicomponent mixture with account for viscous
effects, thermal conductivity, multicomponent diffusion and energy release from chemical transformations. Transport coefficients were evaluated from the first principles of gas kinetics theory [13]. Thermodynamic quantities were calculated from standard thermophysical tables using high-order polynomial interpolation [14]. Chemical reactions were modeled with chemical kinetic scheme of hydrogen oxidation from [13]. Numerical solution of the governing system was obtained with euler-lagrange method [15] modified to ensure stability and second-order in space precision of the numerical solution procedure.

To study the influence of mixture concentration spatial non-uniformities on the flame propagation, calculations were performed with initially defined deviations of hydrogen and oxygen concentration from the mean value. Deviations of the hydrogen concentrations $C_{H_2}$ from the mean, mostly stoichiometric condition were calculated as random quantities, normally distributed with defined dispersion equal to $\Omega$. In order to perform detailed analysis of concentration non-uniformities influence on the combustion process, computations were carried out in two setups. In first setup dependence between the flame propagation speed and dispersion of the concentration deviation was analyzed. In this case small rectangular areas with certain size were randomly picked inside computational domain. In each area initial concentrations were defined with certain dispersion varied for each calculation. For every value of dispersion a set of calculations were performed (from 3 to 6 calculations in each set) with independently generated random deviations. For further analysis results acquired in each set were averaged. Obtained results allowed us to plot a relation between characteristics of flame non-stationary evolution and concentrations dispersion. The second setup was aimed to obtain dependence between the flame propagation speed and concentration non-uniformities spatial scales. In this case the size of rectangular areas was varied from one calculations set to another, while the value dispersion of concentrations deviation in these areas was fixed. On the basis of calculations the dependence between combustion process development parameters and spatial scales of concentration non-uniformities was acquired. Presented results were also averaged for each set of calculations.

Combustion process was modeled inside semi-opened channel of 5 mm width and 100 mm length with adiabatically isolated non-slip walls. Non-slip boundary conditions imposed on the side walls allowed to take into account affect of boundary layer. Channel was filled with hydrogen-oxygen mixture at 1 atm pressure and 300 K temperature. In contrast to [12] components concentrations were non-uniform and only average values were correspond to stoichiometric conditions. Ignition took place near left end-wall of the channel in thin heated layer of 1 mm width. Main results were obtained with high resolution on fine computational grid with cell size 0.05 mm x 0.05 mm, which allowed to precisely reproduce deflagration-to-detonation process in hydrogen-oxygen mixtures [16].

3. Results and discussion
Both experimental and theoretical studies [12, 17] show that the flame front in the premixed stoichiometric hydrogen-oxygen mixture propagates through the semi-opened channel with acceleration up to the sonic speed in the fresh mixture and then the mechanism of transition to detonation switches on. Herewith the evolution of combustion wave passes four basic stages. The first stage represents formation of a stable flame front in the vicinity of ignition region that is fully determined by the characteristics of the ignition source. On the second stage flame propagates with acceleration moving with the flow of the fresh mixture pushed by the expanding combustion products. As the flow is dragged in the boundary layer region and as the flame front itself is subjected to hydrodynamic instability front surface grows and becomes curved. As the flame evolution transits to the third stage the flame structure stabilizes under the influence of nonlinear factors and predomination of long wavelength perturbations. Flame evolution achieves fourth stage once the flame front accelerates up to the sonic speed. On this stage the flame propagation is defined by the peculiarities of combustion in the supersonic flow
that results in formation of sufficiently strong shock wave on the scales of the flame front that in turn leads to the onset of detonation.

Figure 1. Evolution of the flame front propagating through the channel filled with poorly premixed hydrogen-oxygen mixture. Subsequent positions of the flame front are shown with equal time intervals equal to 10 us. (a) – fully premixed stoichiometric mixture; (b) – \(\delta=0.25\) mm; (c) – \(\delta=0.5\) mm; (d) – \(\delta=1.25\) mm; (e) – \(\delta=2.5\) mm; \(\Omega=0.21\).

Figure 2. History of the flame speeds in different poorly premixed mixtures. 1 fully premixed stoichiometric mixture; 2 – 0.25 mm; 3 – 0.5 mm; 4 – 1.5 mm; 5 – 2.5 mm; \(\Omega=0.21\).

Consider now the influence of spatial non-uniformities in the distribution of combustible mixture components on the flame speed. We will analyze the flame propagation in the mixtures with different degrees of stirring from those almost premixed and characterized by small-scale perturbations to those less premixed and characterized by large-scale perturbations. This should allow us to understand the effect of non-uniformities spatial scales on both acceleration and deceleration of the flame front on different stages of flame evolution. Results of numerical analysis show that the spatial non-uniformity in concentrations of reacting mixture components could affect the combustion on all the stages of its evolution. Thus on the first stage of flame initiation the non-uniformity determines the topology of the forming flame front. In turn the formed spectrum of the perturbations defines the initial conditions for evolution of flame front instability, which plays a great role in further evolution of the flame front and its interaction with the flow. Figure 1 illustrates how the degree of concentration spatial non-uniformity affects
the evolution of the flame front in the process of its propagation through the channel filled with poorly premixed mixture. Herewith one can clearly see that the role of small-scale non-uniformities ($\delta=0.25$ mm and $\delta=0.5$ mm in Fig. 1) occurs to be much more significant compared with large-scale ones ($\delta=1.25$ mm and $\delta=2.5$ mm in Fig. 1) that as a result causes faster flame propagation on the second stage of flame evolution in channel. This can be explained by the fact that the initial linear stage of flame front instability is mainly determined by the evolution of short-wavelength perturbations. The presence of small-scale non-uniformities in the flow and their continuous influence on the flame front on the second stage of the process causes saturation of the spectrum of flame front perturbations with short wavelengths. On the following nonlinear stage the energy is transferred from these short wavelengths to the longer ones that in turn provides additional acceleration of the flame front on the third stage of the process. This can be clearly seen from figure 2 representing the histories of flame speeds in poorly premixed mixtures characterized by the same dispersion of concentration and different spatial scales of the non-uniformities. Fig. 2 illustrates a notable increase in acceleration rate in case of poorly premixed mixtures with small-scale non-uniformities ($\delta=0.25 – 1.00$ mm) compared with the case of premixed stoichiometric compound. With the increase in spatial scales of non-uniformities the factor of concentration deviation from the stoichiometric one becomes to play a greater role than the factor of flame front perturbations generation. Due to this the overall process of flame acceleration is alike to that in non-stoichiometric hydrogen-oxygen mixtures where the onset of detonation arises on larger spatio-temporal scales.

To get more comprehensive understanding of the role of chemical composition non-uniformities in the processes of hydrogen-oxygen flame acceleration and DDT let consider the dependence of pre-detonation length (the distance passed by the flame front before onset of detonation) on the non-uniformities characteristics (see. Fig. 3). Figure 3a shows that the dependence of pre-detonation length (and therefore of corresponding time scale) on the non-uniformities scales is non-monotonic. The minimum in the dependence corresponds to $\delta \approx 0.25 – 0.50$ mm that by the order of magnitude corresponds to the theoretical estimate for critical wavelength $\lambda_c$ below which hydrodynamic instability of the flame front (Darrieus-Landau instability) does not evolve. According to the estimate proposed in [18] and taking into account the experimental data, empirical estimations and numerical results for stoichiometric hydrogen-oxygen ($\nu/U_f \approx 10^{-5}$ m, $\theta \approx 7.3$) one can obtain $\lambda_c \approx 0.25$ mm. Perturbations of smaller scales
are not directly involved in the evolution of hydrodynamic instability of the flame front. However their interference on the flame surface as well as the expansion of the flame front perturbed in such a way determine excitation of the modes with wavelengths $\sim \lambda_c$. The perturbations generated by the spatial non-uniformities of larger scales have weaker influence on the Darrieus-Landau instability evolution as the rate of their increase is smaller. Furthermore as it was already mentioned above, with the increase in non-uniformity spatial scales the chemical factor becomes to play a greater role that determines local flame deceleration and as a result integral deceleration of the whole process. The longer process the higher influence of convection and diffusion on the flow structure. As a result the influence of spatial non-uniformities on the structure of the flame surface decreases with time. Due to this the effect of dispersion in mixture composition on the flame evolution is limited in time and decreases with the flame acceleration.

One more factor affecting the scales of DDT is the dispersion value, which characterize the deviation of mixture composition from the average one inside the single non-uniformity. At the fixed value of the non-uniformity spatial scale the increase in dispersion value leads to the increase in amplitude of flame surface perturbations that in turn causes increase in the acceleration rate and faster onset of detonation. However further increase in the deviation of the mixture composition from the stoichiometric one leads to local flame deceleration when entering the regions containing non-stoichiometric mixture. As a result the average flame deceleration can be observed (see Fig. 3b).

In case of averagely non-stoichiometric compositions the non-uniformity in fuel concentration distribution affects the flame surface evolution in the similar way as in stoichiometric case discussed above. Small-scaled non-uniformities lead to the increase in detonability. Figure 4 represents the decrease in pre-detonation length related with the non-uniform composition of lean hydrogen-oxygen mixtures. As one can see the higher rate of acceleration of lean flames is achieved due to the small-scale perturbations just as in the case of stoichiometric mixture. At the same time the effect of perturbations of larger scales weakens with the decrease in average hydrogen concentration and already in equimolar mixture the non-uniformities of $\delta=1.25$ mm cause no additional acceleration of DDT. Presented results indicate a possibility of qualitative changes in the detonation onset predictions for mixtures occupying a certain volume since the estimate of pre-detonation length can vary depending on the degree of stirring. In poorly premixed mixture the pre-detonation length can be significantly shorter than in fully premixed mixture with the same average composition. It can be clearly illustrated on the example of mixture with 45% content of hydrogen. In this case we did not observe DDT in the semi-opened channel of 100 mm length filled with fully premixed mixture. However in case of poorly premixed mixture characterized by small-scaled non-uniformities in hydrogen concentration we observed DDT already at the distance of 68 mm.

On the final fourth stage of flame acceleration on which the flow velocity achieves the sonic speed the physicochemical processes evolving on the scales of reaction zone become to play the leading role [16]. Depending on the mixture composition the process could evolve in two paths: (1) the additional acceleration of the flame front takes place due to the additional compression inside the reaction zone by the chocked compression waves; (2) the regime of flame propagation stabilizes near the sonic speed in the combustion products when the compression rate is not enough to accelerate combustion wave. Thereby in case of non-uniform concentration distribution ahead the flame front the subsequent burning of volumes containing mixtures of different compositions could cause changing in the regimes of supersonic combustion wave. Depending on the dispersion value the DDT can be either accelerated or prevented. After the formation of stable detonation wave the non-uniformities could still affect the process, leading to the disturbance of the reaction zone and as a result to the instability of detonation front.
4. Conclusions
Considered here effect of flame acceleration due to the perturbations in mixture concentration formally can be compared with the flame acceleration related with the flow velocity fluctuations in the turbulized flow. Both factors are intrinsic to the scenarios of accidental hydrogen explosion and are of importance for estimations of the hazardous impact of the explosion. At the first sight the character of influence of both factors should be similar however the quantitative effects are different. In case of flow turbulization the increase in perturbations magnitude (up to the certain limit) leads to the more intensive effect on the flame acceleration. In case of mixture composition perturbations there are two main opportunities. In case of small-scale non-uniformities the increase in perturbations magnitude favors the increase in flame acceleration rate and corresponding decrease in pre-detonation length. Herewith the role of active centers (regions with near stoichiometric mixture composition) is greater than the role of neutral centers (with significant deviation from stoichiometry). On the contrary in case of large-scale non-uniformities the effect of local flame deceleration becomes predominant due to the greater role of neutral centers. Therefore deceleration of DDT process takes place and even the prevention of DDT becomes possible. On the base of distinguished peculiarities one can conclude that the specific feature of considered here instability is its maximal development already at relatively small deviations in the perturbed parameter (fuel concentration) from its average value. In practice the possible presence of weak concentration perturbations in the nearly premixed combustible mixture (that corresponds to the case of small-scale non-uniformities with small dispersion value) their affect on the flame acceleration and DDT could lead to significantly different results from those obtained via prognostic calculations considering combustion of fully premixed mixture.

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