



distributions of temperature and/or mixture concentration.

Thus, phenomenologically, a realistic self-ignition process reveals the following features:

- it proceeds inhomogeneously (in hot spots)
- it's propagation is oriented
- its propagation velocity is time dependent
- a spectrum of propagation velocities is continuous
- it is accompanied with gasdynamic effects

In a classical theory of thermal and chain explosions these features are not taken into account. A combustible mixture is assumed uniform with local temperature and species concentration gradients being neglected. However, in many applications the above features may become important.

Zeldovich et al [25] were the first who studied spontaneous flames theoretically. At present there exists a voluminous literature on theoretical study of spontaneous combustion regimes both in homogeneous [1,4-6,9-12,15,23-25] and heterogeneous [7,11,16] media.

The most important consequence of spontaneous flame origin and propagation is a possibility of strong shock or detonation initiation. This may become a case when the following two requirements are met:

- a reactive mixture is preconditioned for localized self-ignition
- a proper ignition delay gradient is formed in the vicinity of an ignition site*

The latter condition is required for ensuring coupling between spontaneous flame and accompanying pressure waves. Coupling results in shock wave and, subsequently, detonation wave formation [11,26].

Zeldovich et al [26] have classified explosion processes arising spontaneously in a reactive mixture with concentration or temperature nonuniformities. They distinguished the following types of explosion evolution:

- 1) spatially uniform constant volume explosion;
- 2) direct detonation initiation. Detonation arises gradually inside or outside a nonuniformity and propagates steadily in a rest of mixture;
- 3) detonation initiation with a shock precursor. Detonation arises outside a nonuniformity behind a shock precursor generated at earlier stages of explosion;
- 4) oscillatory explosion processes. A reactive shock wave forms inside a nonuniformity and decays outside it giving rise to secondary explosions and secondary shock waves.

Zeldovich et al [26] have shown also that there exists a critical (minimum) size of a nonuniformity for a given mixture which makes it

possible direct detonation initiation. The study of Zeldovich et al [26] was based on a simplified approach making use of a one-step Arrhenius type reaction and neglecting transport processes.

The aim of this paper is to generalize the classification of Zeldovich et al [26] by studying gasdynamic patterns arising in nonuniformly preheated hydrogen-oxygen mixture with due regard for detailed chemical kinetics and transport processes.

## 2. FORMULATION

Consider a volume of dimension  $L$  containing initially quiescent stoichiometric  $H_2 - O_2$  mixture at pressure  $p_0$ . A spatial domain of characteristic dimension  $X_T$  with nonuniform temperature distribution  $T(x)$  is assumed to exist in the volume. For modeling wave patterns generated in the system due to the nonuniform initial conditions we adopt the following assumptions:

- the problem is treated one-dimensionally;
- the combustion is laminar;
- the combustion gases obey the perfect gas law;
- the volume is confined by adiabatic, non-catalytic, impermeable wall

The basic equations used for solving the problem in LaGrangian formulation are as follows

$$\frac{\partial p}{\partial t} + \rho^2 \frac{\partial w}{\partial \psi} = 0; \quad \frac{\partial y_j}{\partial t} + \frac{\partial p y_j v_j}{\partial \psi} = \frac{\dot{\omega}_j}{\rho}, \quad j = 1, \dots, N$$

$$\frac{\partial T}{\partial t} - \frac{1}{\rho c_p} \frac{\partial T}{\partial \psi} + \frac{1}{c_p} \sum_{j=1}^N p y_j v_j c_{pj} \frac{\partial T}{\partial \psi} - \frac{1}{c_p} \frac{\partial}{\partial \psi} (\lambda_T \rho \frac{\partial T}{\partial \psi}) =$$

$$\frac{4}{3} \frac{\rho \eta}{c_p} \frac{\partial w^2}{\partial \psi} + \frac{1}{\rho c_p} \sum_{j=1}^N \dot{\omega}_j h_j = 0 \quad (1)$$

$$\frac{\partial w}{\partial t} + \frac{\partial p}{\partial \psi} - \frac{4}{3} \frac{\partial}{\partial \psi} \left( \rho \eta \frac{\partial w}{\partial \psi} \right) = 0$$

$$p - R^0 \rho T \sum_{j=1}^N y_j / m_j = 0; \quad \frac{\partial x}{\partial \psi} = \frac{1}{\rho}$$

where  $0 \leq \psi \leq \psi_{\max}$  is LaGrangian coordinate,  $x$  Eulerian coordinate,  $t$  time,  $\rho$  density,  $w$  mass velocity,  $T$  temperature,  $y_j$  mass fraction of species  $j$ ,  $N$  number of species,  $v$  diffusion velocity of species  $j$ ,  $m_j$

molecular mass of species  $j$ ,  $\dot{\omega}_j$  molar rate of formation of species  $j$ ,  $c_{p,j}$  specific heat of species  $j$  at constant pressure,  $c_p$  specific heat of the mixture at constant pressure,  $\lambda_T$  thermal conductivity,  $\eta$  dynamic viscosity,  $h_j$  specific enthalpy of species  $j$ ,  $R^0$  universal gas constant.

The set of equations (1) is supplemented by initial conditions

$$\begin{aligned} T(0, \psi) &= T^0(\psi); & y_j(0, \psi) &= y_{j0}, & J &= 1, \dots, N \\ p(0, \psi) &= p_0; & w(0, \psi) &= 0 \end{aligned} \quad (2)$$

and boundary conditions

$$\begin{aligned} \frac{\partial T(t, 0)}{\partial \psi} &= 0; & \frac{\partial T(t, \psi_{\max})}{\partial \psi} &= 0 \\ \frac{\partial y_j(t, 0)}{\partial \psi} &= 0; & \frac{\partial y_j(t, \psi_{\max})}{\partial \psi} &= 0 & J &= 1, \dots, N \\ \frac{\partial p(t, 0)}{\partial \psi} &= 0; & \frac{\partial p(t, \psi_{\max})}{\partial \psi} &= 0 \\ w(t, 0) &= 0; & w(t, \psi_{\max}) &= 0 \\ x(t, 0) &= 0; & x(t, \psi_{\max}) &= X_{\max} \end{aligned} \quad (3)$$

where index 0 denotes values at  $t = 0$ .

Temperature distribution  $T(0, x)$  in a nonuniformity is taken monotonous in the form

$$\begin{aligned} 0 \leq x \leq \delta & & T &= T_m \text{ (peak value)} \\ \delta < x < X_T & & \text{cubic polynom interpolating between } T_m \text{ and } T_e \\ X_T < x < L & & T = T_e \text{ (ground value)} \end{aligned} \quad (4)$$

The function  $T^0(\psi)$  is then derived by using the last equation of Eqs.(1).

In order to observe wave patterns outside a nonuniformity we specify  $L \geq 2X_T$ . The reaction mechanism for  $H_2 - O_2$  mixture and transport coefficients are taken from [14]. The problem (1)-(4) is solved numerically by the method described in detail in [23].

### 3. ESTIMATION OF INITIAL CONDITIONS

For estimating initial values of  $T_m$ ,  $T_e$ ,  $p_0$ , and  $X_T$  at which one can expect spontaneous onset and amplification of shock waves in a stoichiometric  $H_2 - O_2$  mixture we make use of the coupling criterion [11,26]

$$u_s \approx u_g \quad (5)$$

Here  $u_s$  is the spontaneous flame velocity,  $u_g$  the characteristic gasdynamic velocity. A local value of  $u_s$  is defined by Zeldovich [27] as

$$u_g = |\text{grad } t_i|^{-1} \quad (6)$$

where  $t_i$  is the local induction time of mixture self-ignition. A characteristic gasdynamic velocity is

$$u_g = (\gamma p_0 / \rho_0)^{1/2} \quad (7)$$

To obtain an explicit relationship between the basic parameters of the problem in the frame of Eq.(5) we use the local induction time written in the form

$$t_i = A [H_2]^a [O_2]^b \exp(E/RT) \quad (8)$$

where  $[H_2]$  and  $[O_2]$  are the molar concentrations of hydrogen and oxygen respectively,  $a$  and  $b$  the exponents,  $E$  the effective activation energy. Combination of Eqs.(6) and (8) gives

$$u_g = [-t_i (E/RT^2) \text{grad} T]^{-1} \quad (9)$$

This expression introduced into Eq.(5) with due regard for Eq.(7) gives an estimate of the temperature gradient:

$$\text{grad} T = -(RT^2/E) [E t_i (\gamma p / \rho)^{1/2}]^{-1}$$

Assuming  $T$  above the autoignition limit at a given value of  $p_0$  (say,  $p_0 = 0.1$  MPa,  $T = 1000$  K [2,21]) one obtains for a stoichiometric  $H_2 - O_2$  mixture [20]:  $\gamma \approx 1.354$ ,  $\rho_0 = p_0 / RT \approx 0.144$  kg/m<sup>3</sup>, where  $\gamma$  is the ratio of specific heats. Ignition delay data of [17] give for the above example  $E = 18240 \pm 2870$  kal/mole, and the induction time about 50  $\mu\text{sec}$  (for the average  $E$  value). Ignition delay data of [3] give for the same example  $E = 17200$  kal/mole,  $t_i = 160$   $\mu\text{sec}$ . As a result,  $\text{grad} T = (750 - 2200)$  K/m. Choosing  $X_T = 1$  cm as a characteristic size of temperature nonuniformity we come finally to the following set of basic parameters:

$$p_0 = 0.1 \text{ MPa}; T_0 = 1000 \text{ K}; X_T = 1 \text{ cm}; T_m \approx (1010 - 1020) \text{ K}$$

A parameter  $\delta$  entering Eq.(4) is used for ensuring adiabatic conditions at the wall. In calculations  $\delta = 0.2$  cm was assumed.

#### 4. RESULTS OF CALCULATIONS

The correctness of the reaction scheme given in [14] was first confirmed by comparing predicted induction times with available measurements. At  $T_0 = T_m = 1000$  K,  $p_0 = 0.1$  MPa the predicted induction time of a stoichiometric  $H_2 - O_2$  mixture is 99.4  $\mu\text{s}$ , which is close to the measured value [3,17].

At very small temperature increments  $\Delta T = T_m - T_0 \leq 1$  K almost constant volume explosion has been observed in calculations with  $X_T = 1$

cm,  $T_e = 1000$  K,  $p_o = 0.1$  MPa.

Figure 1 shows pressure history in the mixture with a temperature nonuniformity of size  $X_T = 1$  cm with  $T_m = 1010$  K,  $T_e = 1000$  K ( $\Delta T = T_m - T_e = 10$  K),  $p_o = 0.1$  MPa. As is clearly seen, a detonation wave arises spontaneously outside the nonuniformity. The detonation wave propagates at the velocity  $\approx 2560$  m/s which is rather close to the thermodynamic value 2519 m/s, calculated by Vasiliev et al [20] for the outlined conditions. A calculated temperature of combustion products  $\approx 3200$  K which corresponds with the thermodynamic value 3136 K, calculated by Vasiliev et al [20]. At time  $t \approx 100$   $\mu$ s the reaction in front of the detonation wave becomes apparent. The overpressure in a shock wave emanating from the nonuniformity is about  $\Delta p_i \approx 0.4$  MPa. Clearly the explosion process under consideration should be attributed to the second type in classification of Zeldovich et al [26]. Similar results were obtained at  $\Delta T = 30$  K other conditions being equal. The overpressure in a shock wave emanating from the nonuniformity increased in the latter case up to  $\Delta p_i \approx 0.6$  MPa.

Further increase in peak temperature up to  $T_m = 1100$  K ( $\Delta T = 100$  K) results in decreasing the shock overpressure at the

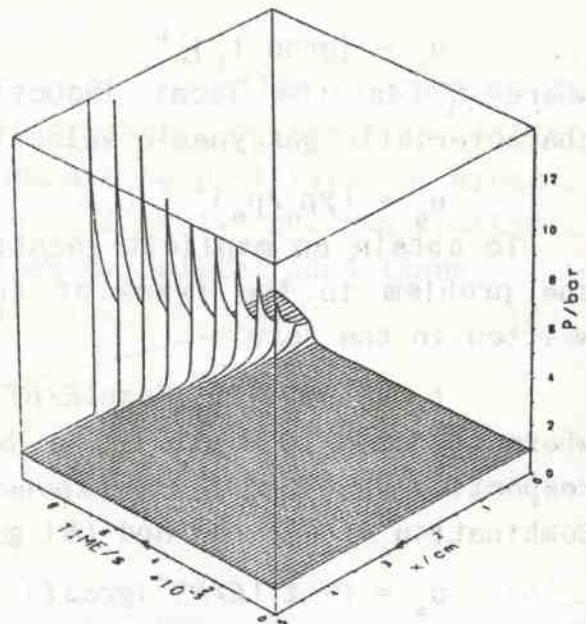


Fig.1 Time-dependent pressure profile in stoichiometric  $H_2-O_2$  mixture with a temperature nonuniformity:  $T_m = 1010$  K;  $T_e = 1000$  K,  $p_o = 0.1$  MPa,  $X_T = 1$  cm.

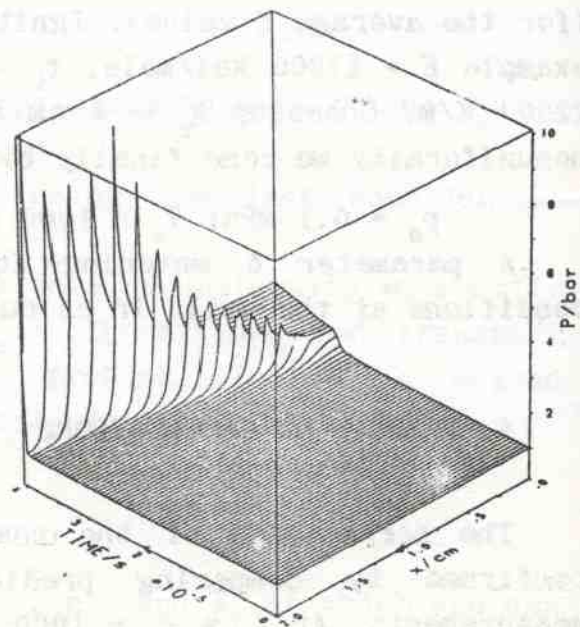


Fig.2 Time-dependent pressure profile in stoichiometric  $H_2-O_2$  mixture with a temperature nonuniformity:  $T_m = 1100$  K;  $T_e = 1000$  K,  $p_o = 0.1$  MPa,  $X_T = 1$  cm.

edge of nonuniformity to  $\Delta p_f \approx 0.4$  MPa (Fig.2). Also evident is the change of explosion dynamics. After emanating from the nonuniformity a shock wave decays. Apparent amplification of the shock wave at  $t \approx 40$   $\mu$ s is a result of its reflection from the opposite wall at  $x = 2$  cm. Shock attenuation is caused by growing difference between reaction induction times at  $T = T_m$  and  $T = T_o$ . The explosion process under consideration should be attributed to the fourth type in the classification of Zeldovich et al [26].

Increasing peak temperature to  $T_m = 1400$  K ( $\Delta T = 400$  K) as in Fig.3 we observe further decrease in shock overpressure to  $\Delta p_f \approx 0.16$  MPa at the edge of the nonuniformity. A reaction front lags behind a shock and propagates in the mode of laminar flame. A shock wave may play the role of shock precursor which preconditions the mixture downstream a flame front.

Therefore the explosion processes of the third type may be expected. Explosion process of the third type was observed numerically in [23,26]. A secondary explosion downstream the flame front, amplification of a secondary shock wave and, finally, the onset of the C-J detonation were detected. The shock overpressure at the edge of a nonuniformity was about 0.1 MPa.

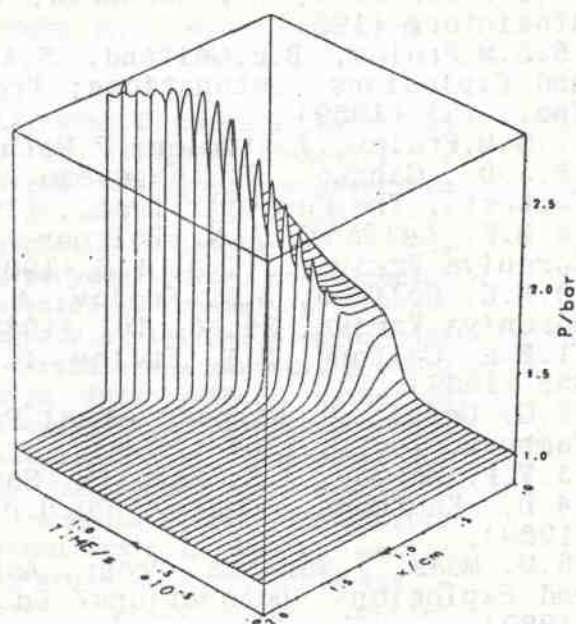


Fig.3 Time-dependent pressure profile in stoichiometric  $H_2 - O_2$  mixture with a temperature nonuniformity:  $T_m = 1400$  K;  $T_o = 1000$  K,  $p_o = 0.1$  MPa,  $X_T = 1$  cm.

## 5. CONCLUSION

The detailed study of explosion dynamics in a nonuniformly preheated mixture confirms the classification of Zeldovich et al [26]. A coupling criterion (Eq.(4)) gives good estimates for predicting the range of parameters giving rise to spontaneous onset of severe explosion processes.

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