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Spontaneous appearance of detonation regime at self-ignition of gaseous mixture: analysis of regularities

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Abstract

The model of spontaneous appearance of detonation regime of distributed exothermic chemical reaction in gaseous mixture is presented. An explanation of the physical mechanism of this phenomenon is based on the concept of collective effects under non-linear wave-kinetic interaction in reacting systems with fluctuations. Dynamics of spontaneous process in reacting system is described assuming uniform macroscopic initial conditions with weak "noise" (uniform in all spectrum of permissible frequencies). Such a statement of a problem allows study general regularities of the phenomenon. The model makes possible to substantiate quantitative criteria of spontaneous deviation of the system from the uniform explosion regime. The results can be used for description of cellular structure of detonation front. The developed approach logically joins the opposite concepts of spontaneous explosion process description: a deterministic approach at the stage of macroscopic non-uniformity development is complemented by a probabilistic one due to using of stochastic infinitesimal disturbances of initial state of the system.

Introduction

The problem of regime of heat explosion is of great scientific interest. Deflagration to detonation transition can be connected with self-ignition of reacting gas at local centers. If the centers with the size exceeding a critical value appear, then one may wait for intensive gasdynamic discontinuities and detonation generation. One of the first this problem was solved in [1] – it was shown that the critical size of the center is determined by condition of balance between heat losses and heat release in the reaction zone.

This mechanism takes place, for instance, at the propagation of laminar flame from the closed end of a tube: the expansion of combustion products is possible only towards fresh mixture, this results in compression wave formation. In this wave the necessary conditions for adiabatic self-ignition of gas can be realized. As it was observed experimentally, the explosion of gas develops in the compression wave, which doesn't yet transform in a shock wave [2, 3]. This means that the detonation wave appears in this case as a result of explosive ignition of adiabatically compressed mixture ahead of the flame front. It is noticeable that the observed detonation wave usually consisted of several perturbations, which indicated the explosion of several volumes of compressed gas at once. This mechanism can lead to local explosive self-ignition phenomena with transition to detonation. According to these ideas the deflagration to detonation transition is the volume explosion originating at local centers in the area ahead of the flame after necessary induction time accumulation ("explosion inside explosion" [4]).

The possibility of spontaneous generation of detonation in spatially non-homogeneous medium was proved for the first time in [5] for the case of non-uniformly distributed temperature using joint numerical solution of the equations of hydrodynamics and chemical kinetics. This research was developed in [6, 7], where the parametric solution of the problem was carried out, the regularities of generation and decay of different types of wave flows were found. Shock-less ignition of strong gasdynamic discontinuities in a medium with non-uniformly distributed pre-exponent factor in one-step reaction of Arrhenius type was studied in [8].

Any direct experiments to study these phenomena were not carried out, but it was recognized in the number of experiments [9-11] devoted to research of self-ignition of gas in adiabatic compression

setups (behind reflected shock waves, or in DDT at obstacles) that self-ignition in the main has local, non-homogeneous character and ignited centers can generate shock waves and initiate detonations. As a result it was offered the conception of "exo-thermal centers of reaction" [12-16]. According to this conception self-ignition of gas is initiated in small areas – exothermal centers of reaction, and gasdynamic flows appearing due to ignition of the centers determine the further development of the process. Partial experimental verifications of these effects can be found in [11, 17, 18].

The phenomenon of spontaneous appearing of shock and detonation waves in the systems at the threshold of self-ignition apparently takes place also in "usual" gaseous detonations. According to [19] the local thermal explosions stochastically arising in the layer of shock-compressed exploding substance behind detonation front play the main role in cellular structure formation and in self-sustained detonation regime providing.

The derivation of a physical model to describe waves in media with heat release on the base of unique evolution equation (for example for pressure) was the aim of works [20, 21] using the approximate description of dynamics of disturbances with small but finite amplitude in a single-phase medium with distributed chemical reaction. With the help of the method of slowly changing wave profile [22] the general system of gasdynamic and chemical kinetics equations for long waves propagating in one direction was reduced to Burgers equation, where the coefficient before the second derivation can change the sign. In the medium with "negative viscosity" the energy pumping effect for small disturbances with certain wavelengths predominates over a dissipation mechanisms. As a result, ignition centers can arise spontaneously in the medium. The possibility for weak shock waves amplification in chemically reacting media was also shown in [23].

On the base of these approaches we fulfilled the complex of works devoted to developing of non-linear model describing evolution of disturbances in such media and allowing to find the condition of transition to detonation wave regime. Developing the previous results, the new model is valid not only for long-wavelength perturbations and describes the evolution of weak stochastic initial plane disturbances propagating in both directions. Due to this the model takes into account the effect of wave collisions, which is of great importance for generation of zones with extremely high reaction rate. Relative simplicity of the created model allows one to predict a qualitative scenario of dynamics of preheated reacting system at the threshold of self-ignition.

Derivation of the evolution equation

Let us consider the evolution of plane waves with small but finite amplitude in homogeneous gaseous medium with non-equilibrium exo-thermal chemical reaction. We'll take into account a finite time of chemical transformation, energy dissipation, non-linearity. The equation of state for gaseous mixture in the general case has the form:

$$\rho = \rho(p,S,Y), \quad \text{or} \quad \frac{d\rho}{dt} \equiv \left(\frac{\partial\rho}{\partial p}\right)_{S,Y}\frac{dp}{dt} + \left(\frac{\partial\rho}{\partial S}\right)_{p,Y}\frac{dS}{dt} + \left(\frac{\partial\rho}{\partial Y}\right)_{S,p}\frac{dY}{dt}, \tag{1}$$

where p is the pressure, ρ is the density, S is the specific enthalpy, t is the time, Y is the mass fraction of reaction products. The reaction is represented in form of one-step transformation «reactants $\rightarrow$ reaction products» with the specific heat release q. The reaction is described by the following kinetic dependence of general form:

$$\omega(p,S,Y) = \rho\frac{dY}{dt}, \tag{2}$$

where ω is the rate of production of reaction products. The expression for entropy production due to non-equilibrium chemical reaction [24]:

$$T\frac{dS}{dt} = -\sum_{j=1}^{2}\mu_j\frac{dY_j}{dt} = q\frac{dY}{dt}, \tag{3}$$

here T is the absolute temperature, μ is the specific chemical potential, subscripts $j = 2,1$ denote reaction products and reactants respectively: $Y_2 \equiv Y$, $Y_1 \equiv (1-Y)$. Taking into account (2), (3), the equation (1) can be written in the form [25, 26]:

$$\frac{d\rho}{dt} = C_f^{-2}\frac{dp}{dt} - F, \tag{4}$$

here $F(p,S,Y) \equiv \overline{q}\omega$, $\overline{q} \equiv \rho\left[\frac{q}{(c_p)_f}\left(\frac{\partial\rho^{-1}}{\partial T}\right)_{p,Y} + \left(\frac{\partial\rho^{-1}}{\partial Y}\right)_{p,S}\right]$, $C_f^2 \equiv \left(\frac{\partial p}{\partial\rho}\right)_{S,Y}$, c_p is the specific heat capacity at constant pressure, C_f is the frozen adiabatic speed of sound in ideal gas. From the equations (1)-(4) we obtain:

$$\left[1+\left(\frac{\tau}{N}\right)_0\frac{d}{dt}\right]\left(\frac{dp}{dt} - \left(C_f\right)_0^2\frac{d\rho}{dt} - \left(C_f\right)_0^2\frac{\gamma-1}{2}\frac{(\rho-\rho_0)}{\rho_0}\frac{\partial\rho}{\partial t}\right) - \delta_0\left(C_f\right)_0^2\frac{\partial\rho}{\partial t} = O\left(\varepsilon^3\right), \tag{5}$$

here $\tau \equiv -\overline{q}\rho F_Y^{-1}$, $L \equiv qF_S\left(T\ F_Y\right)^{-1}$, $N \equiv (1+L)(1-\delta)$, $\delta \equiv \tau F_p C_f^2(1+L)^{-1}$, $C^2 \equiv C_f^2(1-\delta)^{-1}$, ε is the small parameter, $|\tau|$ has the sense of characteristic time of chemical reaction, C is the velocity of wave propagation in «low-frequency» limit, γ is the adiabatic exponent. The equation (5) was first derived in [21]. This equation is dynamic differential equation of state, it describes the dependence of pressure ob gas density in presence of non-equilibrium process, for example chemical reaction or relaxation.

The equation (5) together with continuity equation (6), law of momentum conservation (7) and kinetic dependence (2) forms the closed system describing weak disturbances dynamics in viscous gaseous medium with chemical reaction. The accuracy of the mathematical model is up to the second order of the small parameter.

$$\partial\rho/\partial t + u\,\partial\rho/\partial x + \rho\,\partial u/\partial x = O\left(\varepsilon^3\right), \tag{6}$$

$$\rho\,\partial u/\partial t + \rho_0 u\,\partial u/\partial x + \partial p/\partial x - \nu\rho_0\,\partial^2 u/\partial x^2 = O\left(\varepsilon^3\right). \tag{7}$$

The equations (5), (6) and (7) can be reduced to a single evolution equation for the variable u (or p, ρ). We use the method of slow changing wave profile for the waves propagating in one direction [22]. At the accompanying frame of reference the equation in dimensionless form is written as the following:

$$\left(1 - N_0^{-1}\frac{\partial}{\partial y}\right)\left[2u_z - (\nu\rho)_0 u_{yy} + \frac{\gamma_0+3}{2}uu_y\right] + \delta_0 u_y = 0, \tag{8}$$

here z is the «slow» time, ν is the viscosity.

Study of stability

If we seek the solution of the equation (8) in form of superposition of monochromatic waves: $u = \sum_{j=1}^{\infty} u_j = \sum_{j=1}^{\infty} A_j \exp\left(\Omega_j z + i\kappa_j y\right)$, then we obtain the following dispersion relation:

$$\Omega = \frac{\left(\frac{\delta_0}{2N_0} - \frac{\eta}{2}\right)\kappa^2 - \frac{\eta}{2N_0^2}\kappa^4}{1+\kappa^2/N_0^2} + i\frac{\frac{\beta}{2}\left(1+\kappa^2/N_0^2\right)\kappa^3 - \frac{\delta_0}{2}\kappa}{1+\kappa^2/N_0^2}. \qquad (9)$$

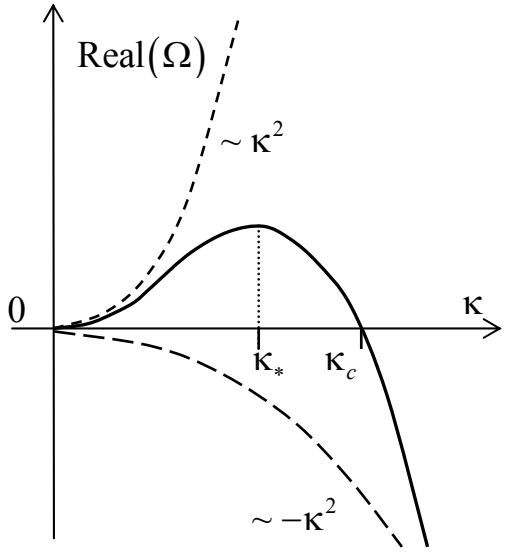

Figure 1. The dependence of increment on wave number.

In the case of short waves ($\kappa \ll 1$) the relation (9) corresponds to linear limit of the Burgers equation describing disturbance attenuation in chemically inertial medium: $\Omega \sim -(\nu\rho)_0\kappa^2$. In the long-wavelength limit $\left(\kappa/N_0\right)^2 \sim \mathrm{O}(\varepsilon)$ the dispersion relation takes into account chemical reaction: $2\Omega = \left(\delta/N - \nu\rho\right)_0 \kappa^2 - i\delta_0\kappa$. This relation also corresponds to linearized Burgers equation with the difference from high-frequency limit: the coefficient before the second derivation can be both positive and negative (see figure 1). If the condition (10) is satisfied

$$\delta/N > \nu\rho\,, \qquad (10)$$

then long-wavelength disturbances will increase because energy pumping due to reaction heat release will exceed energy dissipation. Linear low-frequency waves has the velocity C_0 , which exceeds the frozen speed of sound C_f if $\delta_0 > 0$ (the subscript 0 denotes the undisturbed state).

Numerical modeling

To be solved numerically the equation (8) was approximated with finite differences using implicit two-level central-symmetrical scheme of the second order $\mathrm{O}\left(\hat{\tau} + h^2\right)$. The sweep method and boundary conditions of constant solution at $y \to \pm\infty$ were used. The number of mesh points M=1500. The following values of the parameters were set: $h = 1/3;\ \hat{\tau} = 1/30;\ N_0 = 1/3;\ \delta_0 = 2/3\,;\ (\nu\rho)_0 = 2{,}6$. In the calculations a shock wave was used as the initial condition. Its amplitude was changed within the diapason from $W = 0{,}2$ to $W = 2{,}5$. Shock waves with the initial amplitudes from the interval 0,2-2,0 have different evolution but in each case the solitary wave was finally formed. This wave moves more quickly than initial shock, its parameters don't depend on W: the amplitude is 2,5, the velocity in accompanying frame is 1,8 (see figures 2 and 3). As the initial amplitude increases, the solitary wave appears earlier. If W reaches or exceeds the amplitude of the solitary wave, then self-sustained solitary structure doesn't appear, the oscillations of the leading peak parameters are observed (see figure 4).

The obtained analytical and numerical results allow us to make some conclusions on the physical mechanism of solitary wave formation. It is dissipative auto-structure spontaneously arising as a result

of non-linear wave-kinetic interaction in non-equilibrium system and belongs to the type of spatially non-uniform structures. The energy of long-wavelength perturbations of state parameters increases in the reaction zone due to heat release. Non-linearity leads to high-frequency spectrum component arising. For these disturbances the energy dissipation predominates. As a result the energy is transferred from low- to high-frequency disturbances and the steady-state spectrum is formed, i.e. the stationary dissipative structure appears.

Analysis of regularities of spontaneous detonation regime appearing at self-ignition of medium

At this section the problem of appearing of so-called spontaneous detonation regimes of distributed non-equilibrium chemical reaction in gaseous mixture is considered basing at the approach described above. In the works [5, 27, 6, 28] the conditions for spontaneous arising of detonation in reacting systems were analyzed. A high sensitivity of explosion type to small changing of thermodynamic and kinetic parameters of the medium (for example initial temperature) was found, which is usual for bifurcation points of self-organized systems. The explanation of physical mechanism of this phenomenon (and also derivation of criteria) apparently has to be based at the conceptions of collective effect in non-linear wave-kinetic interaction in reacting systems with fluctuations [29, 30].

Let us consider the spontaneous process dynamics using macroscopically uniform initial conditions with superimposed weak noise (with homogeneous spectrum). Such the statement of the problem allows to study general regularities of the phenomenon, and the gradient mechanism of detonation appearing turns out to be a special case of spontaneous evolution of reacting system. We reduce the system of equations (5), (6), (7) to the single non-linear integral-differential equation (11) without the supposition of slow changing of solution and write this equation in immovable coordinate frame. This equation describes plane wave propagation in both directions:

$$\left[1+N_0^{-1}\frac{\partial}{\partial t}\right]\left\{\left(\frac{\partial^2}{\partial t^2}-\frac{\partial^2}{\partial x^2}\right)u-(\nu\rho)_0\frac{\partial^3 u}{\partial x^2\partial t}+\right.$$
$$\left.+\frac{\partial}{\partial x}\left(u\frac{\partial u}{\partial t}+\frac{(\gamma_0+1)}{2}\frac{\partial u}{\partial x}\int\frac{\partial u}{\partial x}dt\right)\right\}-\delta_0\frac{\partial^2 u}{\partial x^2}=o(\varepsilon^2), \qquad (11)$$

and the same form have the equations for pressure and for density. The equation (11) coincides with (8), if we assume that the solution depends on $y=x-t$ and $z=\varepsilon t$.

Discussion

The model based on the equations (11) и (2), describes the spontaneous generation of macroscopic non-homogeneities in chemically reacting systems without creation of any special initial conditions with macroscopic gradient of induction time [31-33]. This model describes plane waves propagating in both directions, due to which it takes into account the effect of wave collisions that is of great importance for generation of zones with relatively high reaction rate and increased heat release (i.e centers of explosion). It should be emphasized that is the essence of physical mechanism of self-acceleration of spontaneous generation of macroscopic non-homogeneities with finite amplitude in the initially uniform media with weak stochastic disturbances of parameters of state [34, 35].

Weak stochastic disturbances of uniform initial distribution of mixture density (or reactant concentration) are connected with perturbations of the temperature field. Small fluctuations of temperature causes the remarkable changes of induction time (due to exponential kinetic dependence). If the criterion (10) is locally satisfied then the amplification of weak disturbances (with wave number $\kappa\sim\kappa_*$) takes place and leads to the formation of a set of weak shock waves. These waves collide with each other and generate "spots" with increased values of state parameters. In this way the instability of the system (i.e. the positive wave-kinetic feedback) and the collective interaction between the generated non-homogeneities result in the non-uniform (detonation) regime of explosion.

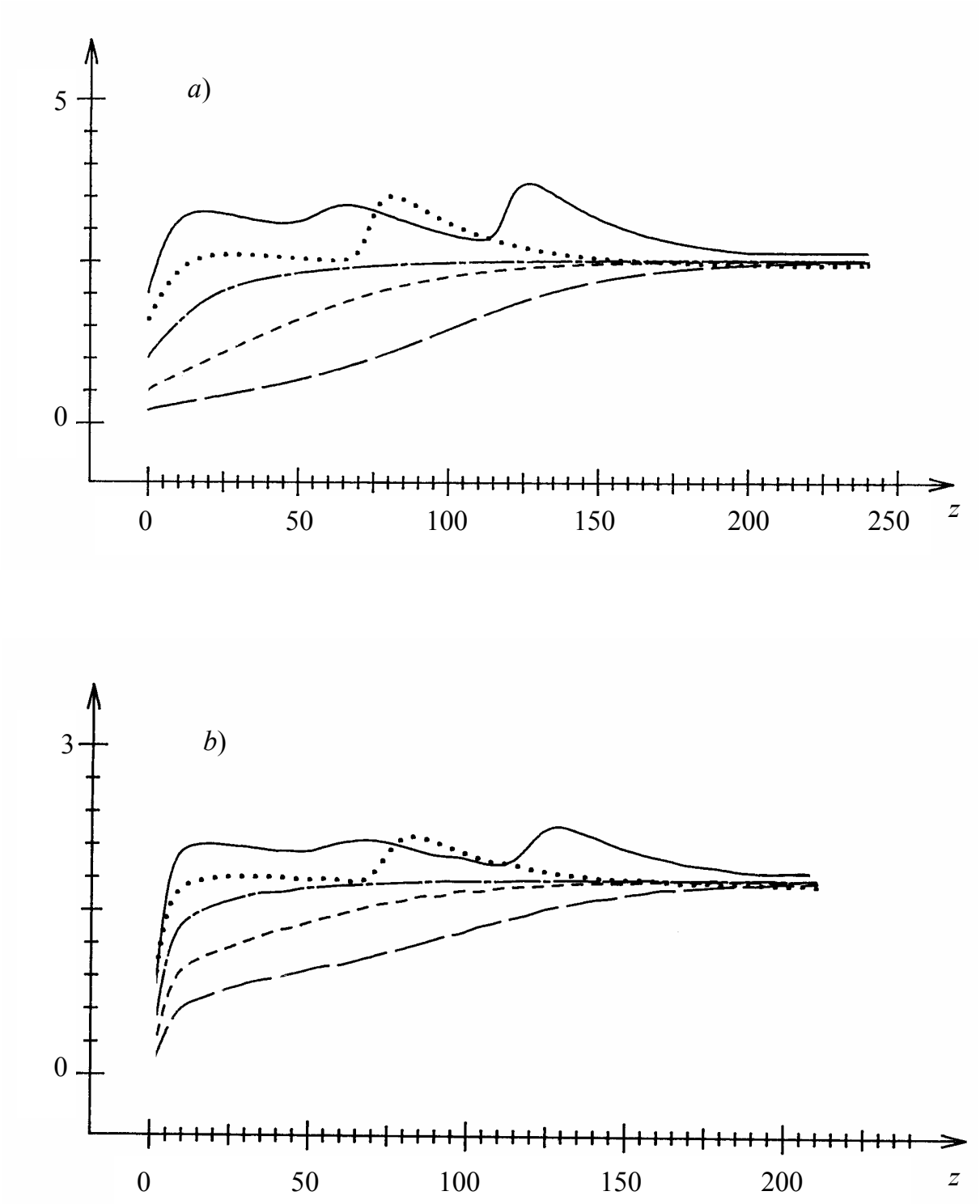

Figure 2. Time dependences of the amplitude (a) and velocity (b) of leading peak at the different values of initial wave amplitude: W=0.2; 0.5; 1.0; 1.5; 2.0.

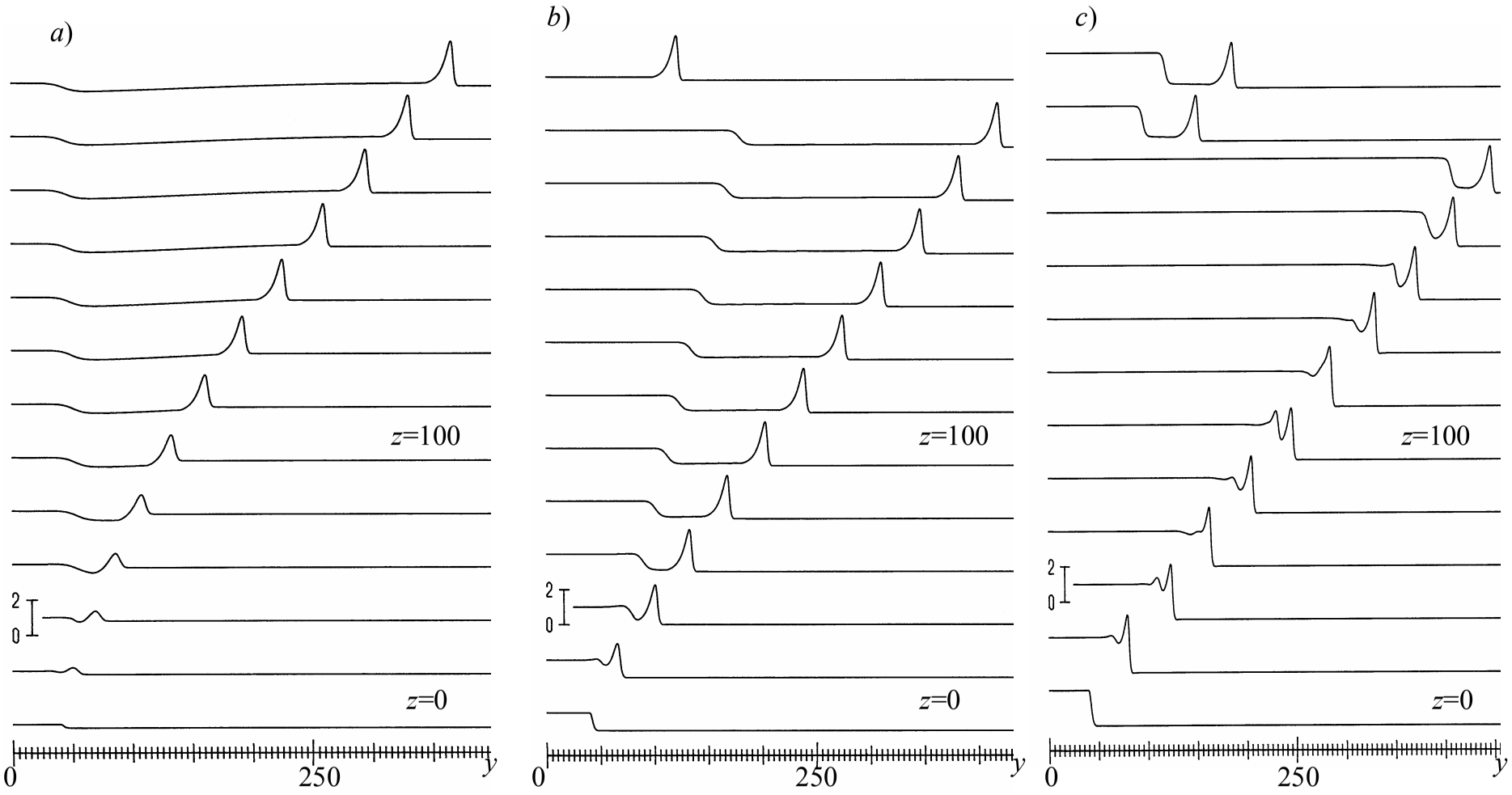

Figure 3. Evolution of the waves with different initial amplitudes: *a*) W=0,2; *b*) W=1,0; *c*) W=2,0. Time step equals to 20.

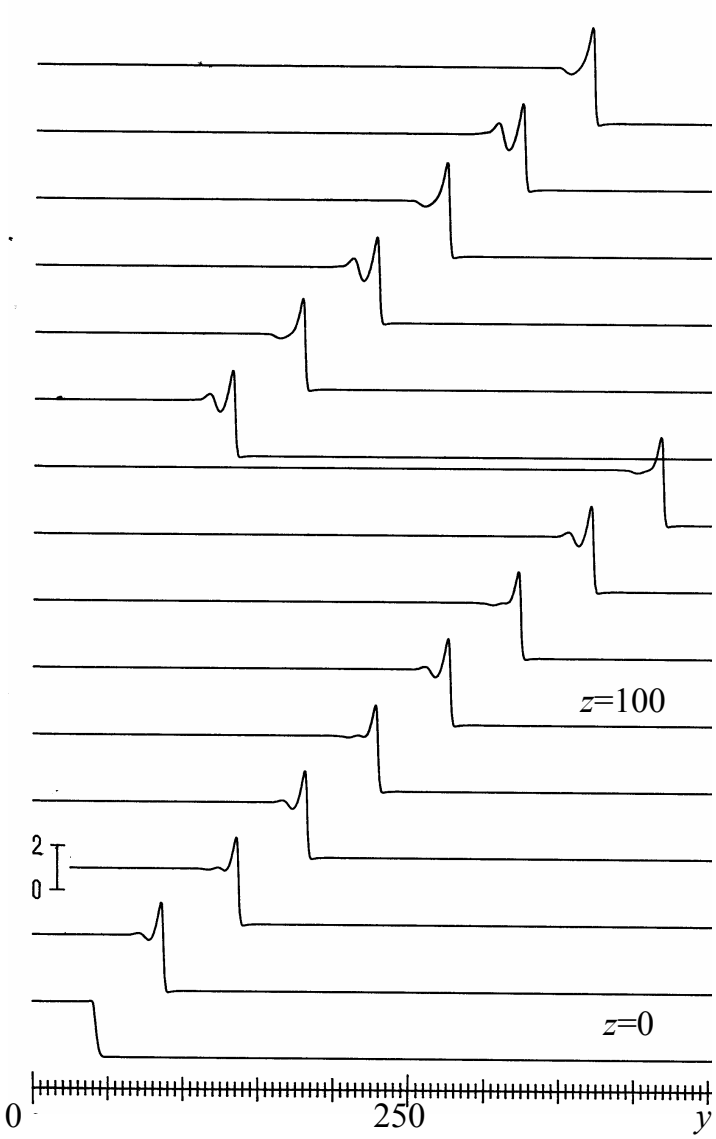

Figure 4. Wave evolution at initial amplitude *W*=2,5. Time step equals to 20.

This mechanism does not need any initial gradient of induction time. This conclusion can be proved by thorough analysis of the results of direct calculations [36] carried out for the case of initial conditions with temperature or concentration gradients. This calculations show that characteristic wavelength $\Lambda_* = 2\pi/\kappa_*$ of amplified pressure of temperature wave at the initial (quasi-linear) stage doesn't depend on the spatial size of superimposed initial non-homogeneity Λ_{in}. In all numerical experiments the detonation regime was observed as $\Lambda_{in} \geq \Lambda_c = 2\pi/\kappa_c$. But the character of the process changed abruptly if $\Lambda_{in} < \Lambda_c$. The detonation did not arise: after the time about τ_0 the uniform thermal explosion occurred. The absence of detonation in this case is connected with the artificial elimination of unstable long-wavelength disturbances from the initial disturbance spectrum. A nonlinear collective interaction of non-homogeneities was not considered in calculations [36], but one can wait that it is able to lead to detonation regime (under certain conditions) even in the case when the unstable harmonics ($\Lambda > \Lambda_c$) are suppressed significantly at the initial time.

Conclusion

The presented model describes the propagation of plane waves developing from infinitesimal (sound) disturbances to weak non-steady shock waves due to non-linearity and positive feedback between

changing of the parameters of state and the rate of reaction heat release. Such the behavior of the solution corresponds to the phenomenon of spontaneous generation of detonation explosions in the systems at the threshold of self-ignition. The model takes into account the energy dissipation and this allows us to formulate the quantitative criteria of spontaneous deviation of the system from the uniform explosion regime: (a) chemical reaction is far enough from equilibrium (the condition (10) governing the amplification of weak long-wavelength disturbances); (b) the size of the system exceeds the wavelength of neutrally stable disturbance Λ_c . These criteria explain the success of the phenomenological conceptions, which give the main role to the geometrical size of initial non-homogeneity (the size of the "germ" or ignition center) in the spontaneous generation of the detonation regime. The account of propagation of disturbances in the opposite directions and collisions of waves allows to simulate a sharp local acceleration of the reaction and shock wave formation. Thank to this the generation of the detonation regime can be described without using of a special macroscopic initial distribution of the temperature (concentration). The developed approach logically joins the opposite concepts (see [36] and [37]) of spontaneous explosion process description: a deterministic approach at the stage of macroscopic non-uniformity development is complemented by a probabilistic one due to using of stochastic infinitesimal disturbances of initial state of the system.

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