The Model of Generation of Spontaneous Explosion (Detonation) Process

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Abstract

The model of dynamics of small but finite-amplitude perturbations in macroscopically uniform chemically non-equilibrium systems with fluctuations close to self-ignition is substantiated. The criterion of stability loss is formulated for uniform state of a system. The conditions of transition from the thermal explosion regime to the detonation regime of chemical transformation are studied. On the base of concept of collective effects in non-linear wave-kinetic interaction the fundamental regularities of spontaneous detonation process generation and development are analyzed.

Introduction

The reacting systems belonging to so-called systems of synergetic type (i.e. systems possessing the property of self-organization) means the existence of principle criteria determining the conditions of changing (bifurcation) of process types differing by dynamics and spatial symmetry (in particular, the conditions of transition from the uniform thermal explosion regime to detonation-wave one). Any model created for explanation and description of the processes in these systems is based at the fundamental Rayleigh principle consisting of the possibility of amplification for weak stochastic perturbations when a positive feedback presents (for example, the feedback between reaction heat release and pressure [1]). In contrast to the description of detonation wave dynamics using the model of ideal reacting continuous medium the investigation of generation stage of wave explosion process must take into account the effect of energy dissipation. The level of energy losses together with the degree of irreversibility of exothermic chemical reaction determine a possibility of spontaneous generation and development of non-uniformities which are ignition centers in the medium.

It was shown that weak (about 1%) initial perturbations of temperature (concentration) uniform field of the system close to self-ignition can result (depending on the spatial size of non-uniformity) in various regimes: constant volume explosion with uniform pressure raising, self-sustained Chapman-Jouguet detonation regime, non-stationary detonation with a forerunner, set of shock waves. At the same time it turns out [2] that as the collective wave interaction is taken into account, the sizes of non-uniformities are not the governing factors with respect to regime of the process. The analysis of known results of numerical modeling allows to conclude that the spatial size and amplitude of non-uniformities in initial parameters don't have a direct relation to fundamental regularities in generation and development of spontaneous explosion (detonation) processes in systems at the threshold of self-ignition. The explanation of physical mechanism of this phenomenon (as well as the formulation of regime changing criteria) apparently is to be built on the base of a concept of collective effects under the non-linear wave-kinetic interaction in reactionable systems with fluctuations. The numerical modeling of so-called gradient mechanism of detonation generation [3] demonstrates a possibility of formation of waves propagating in the direction determined by initial data. It was shown that under certain conditions the process obtains the character of detonation one. For this situation the main governing parameter is the value of temperature gradient (i.e. induction time gradient) in the medium which is preassigned as the initial data. It is well-known that in problems of a spatial structure spontaneous formation a relatively weak variation in parameters of the system can significantly change a solution (not only quantitatively but even qualitatively). This is the general property of incorrect (in Hadamard sense) mathematical problems and physical systems with instability. One can conclude that the regimes modeled in [2, 3] are not rigorously spontaneous because they need the creation of special (artificial) initial conditions. In this respect the introduced in [4] term "spontaneous flame" to denote the process of self-ignition in the medium with non-uniform initial distribution of chemical reaction induction time doesn't seem sufficiently adequate. In the work [5] the other name for this phenomenon is given, namely the "induction flame". In the present work the dynamics of spontaneous processes in reactionable systems is studied beginning with macroscopically uniform initial state with superimposed weak "noise" in all the spectrum of possible frequencies. This statement of the problem allows to study the general regularities of the phenomenon. In this case the gradient mechanism of detonation rise turns out to be the special case of spontaneous evolution of reacting system.

The statement of the problem

The process of perturbation propagation (evolution) at a chemically active gaseous mixture is analyzed. Wave disturbance of uniform state of medium compresses and adiabatically heats the gas, accelerates an exothermic reaction. We have analyzed only one-dimensional case because i) modeling of multi-dimensional flows is very complicate even for contemporary computers, ii) one-dimensional approach is informative enough for qualitative description and explanation of main real physical mechanisms of the processes at self-organizing systems like the analyzed one (for example, one- and two-dimensional approach provides the important results in the studies of a number of other physical problems [6-8]). One may accept the one-dimensional model as a basis for understanding and explanation of real multi-dimensional processes.

The system of fluid dynamic and chemical kinetic equations is the basis for derivation of a single equation describing the structure and dynamics of small but finite-amplitude perturbations:

$$\left(1+\frac{\bar{\tau}}{N}\frac{\partial}{\partial\bar{t}}\right)\frac{\partial}{\partial\bar{t}}\left\{\bar{\rho}(\bar{x},\bar{t})+\bar{\eta}\frac{\partial\bar{\rho}(\bar{x},\bar{t})}{\partial\bar{t}}+\frac{\gamma+1}{4}\bar{\rho}^2(\bar{x},\bar{t})-\right.$$
$$\left.-\int_0^{\bar{x}}\left(\int_0^{\bar{x}_1}\left[\frac{\partial^2\bar{\rho}(\bar{x}_2,\bar{t})}{\partial\bar{t}^2}-2\left(\frac{\partial\bar{\rho}(\bar{x}_2,\bar{t})}{\partial\bar{t}}\right)^2-2\frac{\partial^2\bar{\rho}(\bar{x}_2,\bar{t})}{\partial\bar{t}\partial\bar{x}_2}\int_0^{\bar{x}_2}\frac{\partial\bar{\rho}(\bar{x}_3,\bar{t})}{\partial\bar{t}}d\bar{x}_3\right]d\bar{x}_2\right)d\bar{x}_1\right\}+\alpha\frac{\partial\bar{\rho}(\bar{x},\bar{t})}{\partial\bar{t}}+\mathrm{O}(\varepsilon^3)=0, \quad (1)$$

where γ is the adiabatic exponent of the mixture, $\bar{\rho}$ is the dimesionless perturbation of initial density of the gas mixture, $\alpha^{1/2}$ is the ratio between equilibrium sound speed and "frozen" one, $\bar{\eta}$ is the generalized dissipation coefficient, $\bar{x}$ and $\bar{t}$ are dimensionless coordinate and time, ε is the small parameter ($\varepsilon<<1$). $\bar{\tau}, N$ are dimensionless variables determined by the kinetic equation. $|\bar{\tau}|$ is characteristic reaction time. It is to be emphasized that the equation (1) is derived under the condition of small-amplitude disturbances of pressure, density and velocity, but it doesn't include any assumption on the non-linearity of chemical kinetic law.

Analysis of results

Let us now analyze the equation (1) for the case of waves propagating in one direction. Weak dispersion and dissipation allows one to use the method of slowly changing wave profile in a wave-following coordinate frame: $y=\bar{x}-\bar{t},\xi=\varepsilon\bar{t}$ (ξ is the "slow time"):

$$\frac{\partial}{\partial\bar{x}}=\frac{\partial}{\partial y};\frac{\partial}{\partial\bar{t}}=\frac{\partial}{\partial\xi}\frac{\partial\xi}{\partial\bar{t}}+\frac{\partial}{\partial y}\frac{\partial y}{\partial\bar{t}}=\varepsilon\frac{\partial}{\partial\xi}-\frac{\partial}{\partial y}=\frac{\partial}{\partial z}-\frac{\partial}{\partial y},$$

where $\frac{\partial}{\partial z}\sim\mathrm{O}(\varepsilon)$. Then equation (1) can be transformed to the following

$$\left(1-\frac{\bar{\tau}}{N}\frac{\partial}{\partial y}\right)\left\{\frac{\partial\bar{\rho}}{\partial z}-\frac{\bar{\eta}}{2}\frac{\partial^2\bar{\rho}}{\partial y^2}+\Psi\bar{\rho}\frac{\partial\bar{\rho}}{\partial y}\right\}+\frac{\alpha}{2}\frac{\partial\bar{\rho}}{\partial y}+\mathrm{O}(\varepsilon^3)=0, \quad (2)$$

where $\Psi=1+\frac{\gamma+1}{4}$ is the total coefficient of fluid-dynamic and thermodynamic non-linearities.

To analyze the linear properties of equation (2) let us eliminate in it the fluid-dynamic, thermodynamic and chemical non-linearities: $\Psi=0,\bar{\tau}=1,N=\mathrm{Const},\alpha=\frac{C_0^2}{C_{f0}^2}-1=\mathrm{Const}$. We'll seek a solution in the form of superposition of non-interacting harmonics: $\bar{\rho}_j=\sum_{j=0}^{\infty}r_j\exp(\Omega_j z+ik_j y)$. This leads to the dispersion relation:

$$\Omega=\frac{\left(\frac{\alpha}{2N}-\frac{\bar{\eta}}{2}\right)k^2-\frac{\bar{\eta}}{2N^2}k^4}{1+\frac{k^2}{N^2}}-i\frac{\frac{\alpha}{2}k}{1+\frac{k^2}{N^2}}. \quad (3)$$

High-frequency limiting case

As $k^2=\mathrm{O}(\varepsilon^{-1})$ the reaction is "frozen", then equation (3) is reduced to the following:

$$\Omega \to -\frac{\overline{\eta}}{2}k^2. \tag{4}$$

Relation (4) corresponds to the linear limit of the well-known Burgers' equation which describes the propagation of waves in a chemically inert dissipative medium. According to (4) linear disturbances travel through the medium with the "frozen" sound speed C_{f0} and are attenuated due to energy dissipation.

Low-frequency limiting case

As $k^2 \sim \mathrm{O}(\varepsilon)$ the chemical reaction significantly effects on the evolution of linear disturbances. Instead of (3):

$$\Omega = \left(\frac{\alpha}{2N} - \frac{\overline{\eta}}{2}\right)k^2 - i\frac{\alpha}{2}k. \tag{5}$$

Relation (5) also corresponds to the linearized Burgers' equation, but the velocity of the wave here equals to equilibrium sound speed and the coefficient before the second derivative $\left(\frac{\alpha}{2N} - \frac{\overline{\eta}}{2}\right)$ can be either negative or positive. The latter implies the possibility of amplification for low-frequency harmonics, which takes place as the effect of reaction heat release predominates over energy dissipation:

$$\frac{\alpha}{N} > \overline{\eta}. \tag{6}$$

The value $\overline{\eta}$ at the present model may be considered as the generalized coefficient of energy dissipation (not only due to viscosity but also thermal conductivity, radiation, etc.).

According to the evolution equation (2) non-linear interaction between harmonics could feed energy from unstable low-frequency disturbances to attenuated high-frequency ones. As a result the stabilization of the solution spectrum is possible. A similar mechanism of stabilization of the solution is known for a number of problems, related to the self-organization theory. As examples we quote here the front structure of a laminar flame, the surface of a liquid film flowing down an inclined wall, the solid surface sublimated by a laser radiation, the front structure of a weak alloy solidification, multi-headed gaseous detonations, and others. All these phenomena are followed by appearing of so-called dissipative structures which exist in the steady-state regime owing to energy exchange with the medium, and the decay of high-frequency harmonics is caused by various physical mechanisms: thermal conductivity, surface tension, viscosity, etc.

Discussion

The model (1) in contrast to the previous results [9-11] doesn't include the assumption of the slow changing of solution (i.e. wave profile) in time, thus it is suitable for simulation of a sharp (explosive) change of local values of medium state parameters.

The model allows the possibility of amplification for low-frequency harmonics, which takes place as the effect of reaction heat release predominates over energy dissipation. According to the evolution equation non-linear interaction between harmonics could feed energy from unstable low-frequency disturbances to attenuated high-frequency ones. As a result the stabilization of the solution spectrum is possible.

In spite of the relative simplicity of the non-linear equation, we have failed to find its exact analytical solution (even in the case $N = \mathrm{Const}, \alpha = \mathrm{Const}$). Analytical solutions are known only for essentially more simple equations with given kind of non-linearity. So the further investigation of its solutions needs in using of numerical modeling methods, which have been carried out. It was numerically modeled the quasi-linear process of spontaneous formation of quasi-stationary solitary auto-wave (detonation wave) without the creation of a special initial conditions with macroscopic gradient of induction time.

Conclusions

In conclusion it may be noted that the derived model describes the propagation of planar waves in the system evolving from infinitesimal (sound) perturbations up to weak non-stationary shock waves due to non-linearity and positive feedback between the change of state parameters and the rate of reaction heat release. Such behavior of solution qualitatively corresponds to the phenomenon of detonation explosion spontaneous generation in reacting systems close to the self-ignition threshold. Taking the energy dissipation into account this model allows to formulate specific quantitative criteria of spontaneous deviation of the system from the regime of uniform explosion: a) the flowing chemical reaction must be sufficiently far from the equilibrium conditions (as this condition (6) is true the weak long-wavelength perturbations turn out to be increasing with time); b) the

size of the system must exceed the critical wavelength. These criteria make possible to explain the success of phenomenological conceptions assigning the main role in detonation regime spontaneous generation to the geometric size of initial non-uniformity of parameters of the system (i.e. to the size of a “germ” or “point” of ignition). Taking account of collisions of the waves coming from the opposite directions allows to simulate on the basis of the proposed model a sharp local acceleration of reaction and formation of strong shock waves. As a consequence the transition of the system to detonation explosion regime can be described without using of a special macroscopic spatial distribution of initial temperature (or concentration). The last logically joins the opposite conceptions [2, 12] of spontaneous explosive process description: the deterministic approach at the stage of macro-non-uniformities development is complemented by probabilistic approach because of using a stochastic infinitesimal perturbations (“noise”) of the initial state of the system.

The using of the derived model can bring a positive effect in economy of the efforts required for natural modeling and computer simulation of the technological processes because it provides a general understanding of physical essence of these processes and also the compact mathematical formulation and criteria for quantitative estimations.

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