Journal of Technical Physics, J. Tech. Phys., **46**, 3, 129–143, 2005.
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MODELLING OF CONVECTIVE HEAT EXPLOSION

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A strong temperature dependence of the reaction rate can result, for an exothermic chemical reaction, in heat explosion. In this work we study numerically the influence of natural convection on heat explosion. The model consists of the reaction-diffusion equation coupled with the Navier–Stokes equations under the Boussinesq approximation. A new type of heat explosion, the oscillating heat explosion is found. Critical conditions of explosion and complex nonlinear dynamics of the reaction-diffusion-convection problem are studied.

1. Mathematical Models of Heat Explosion

The storage of chemically reacting products represents a fire and explosion hazard due to a possible heat explosion. An exothermic chemical reaction, even if it is very slow at the ambient temperature, can lead to heat accumulation and temperature raise. Since the reaction rate often increases with temperature, the process self-accelerates and can result in essential temperature increase and finally in explosion. The critical conditions of heat explosion are determined by the balance between heat production because of the exothermic reaction and heat loss through the walls.

The first quantitative model of heat explosion is suggested by Semenov [6, 7] and consists of the ordinary differential equation

$$\frac{d\theta}{dt} = K(\theta) - \alpha\theta \tag{1.1}$$

for the dimensionless temperature θ. This model is based on two principal assumptions: the temperature distribution inside the reactor is homogeneous in space, and consumption of the reactant can be neglected. Both assumptions have clear physical meaning, and they have been justified in numerous experimental and theoretical works (see [8]). In particular, the second assumption means that the explosion occurs at the initial stage of the reaction where the change of the reaction rate due to the reactant depletion is not yet essential. This is why the first term in the right-hand side of (1.1), which describes the reaction rate, depends only on temperature and does not depend on the concentration. The second term describes the heat loss through the reactor walls. The function

$K(\theta)$ is often considered in the form of the Arrhenius exponential. Its approximation by the usual exponential

$$K(\theta) = k_0 e^{\theta}$$

simplifies the analysis of the problem.

Depending on the parameters k_0 and α, Eq. (1.1) can have two stationary solutions, only one solution, or not to have them at all. In the critical case where there exists only one solution θ_0, the functions in the right-hand side of (1.1) and their derivatives coincide at $\theta = \theta_0$:

$$k_0 e^{\theta_0} = \alpha\theta_0, \qquad k_0 e^{\theta_0} = \alpha.$$

From these two conditions we can explicitly find the critical temperature $\theta_0 = 1$, and the critical value of the heat loss coefficient, $\alpha_c = k_0 e$.

If α is less than the critical value, then there are no stationary solutions. For any initial temperature $\theta(0)$, the solution increases in time and becomes unbounded in a finite time. In the real physical situation the temperature remains bounded. It can be unbounded in the model because we have approximated the Arrhenius exponential by the usual exponential. However, the difference between them becomes essential only for very big temperatures, and the critical condition of the explosion remains practically the same.

Thus, in the framework of this model we interpret heat explosion as an unbounded temperature increase. It occurs if the coefficient of heat loss is less than the critical one. If α is greater than the critical value, then there are two stationary solutions. For small initial temperatures, the solution converges to the stable stationary solution, if the initial temperature is sufficiently large, then the solution increases and becomes unbounded in a finite time.

The next step in the development of the theory of heat explosion is related to works by Frank–Kamenetskii [4]. He removed the assumption that the temperature was homogeneous in space and described the phenomenon of heat explosion by the reaction-diffusion equation

$$\frac{\partial\theta}{\partial t} = \Delta\theta + K(\theta) \tag{1.2}$$

assuming that the dimensionless temperature equals 0 at the boundary of the domain. As above, critical conditions of heat explosion are considered as a critical value of parameters where the stationary solutions disappear. In the one-dimensional spatial case and also in two- and three-dimensional radially symmetrical cases, critical conditions of the existence of stationary solutions can be found explicitly (see [1]). There exists a critical value of the parameter k_0 such that stationary solutions do not exist for greater values, and the explosion occurs, that is for any initial temperature distribution the temperature becomes unbounded in a finite time. The first works were followed by a large number of physical and mathematical works where various aspects of this model were studied (see [1, 8]).

One of the further directions of the development of the theory of heat explosion concerns the influence of natural convection on it. If the reaction occurs in a liquid or in a gaseous phase, then a nonhomogeneous temperature distribution can lead to convection that would change the temperature distribution and the critical conditions of the explosion. In the two-dimensional spatial case this process can be described by the reaction-diffusion-convection system of equations

$$\frac{\partial \theta}{\partial t} + u\frac{\partial \theta}{\partial x} + v\frac{\partial \theta}{\partial y} = \Delta\theta + K(\theta), \tag{1.3}$$

$$\frac{\partial u}{\partial t} + u\frac{\partial u}{\partial x} + v\frac{\partial u}{\partial y} = -\frac{\partial p}{\partial x} + P\Delta u, \tag{1.4}$$

$$\frac{\partial v}{\partial t} + u\frac{\partial v}{\partial x} + v\frac{\partial v}{\partial y} = -\frac{\partial p}{\partial y} + P\Delta v + R\theta, \tag{1.5}$$

$$\frac{\partial u}{\partial x} + \frac{\partial v}{\partial y} = 0, \tag{1.6}$$

where u and v are the horizontal and the vertical components of the velocity, respectively, p is the pressure, P is the Prandtl number, and R is the Rayleigh number. This model was first studied in [5]. It was considered in a square domain $0 \le x \le L_x$, $0 \le y \le L_y$, $L_x = L_y$ with the no-slip boundary conditions for the velocity and with the boundary conditions

$$x = 0, L_x : \frac{\partial \theta}{\partial x} = 0, \qquad y = 0, L_y : \theta = 0 \tag{1.7}$$

for the temperature. It can be easily verified that the one-dimensional solution of equation (1.2) is also a solution of problem (1.3)–(1.7). For small Rayleigh numbers this solution is stable, and there is no convection. For large Rayleigh numbers this stationary solution loses its stability and stationary convective regimes appear. It is shown that convection can change the critical conditions of explosion.

The same model is studied in [3] where periodic oscillating regimes are found and a new type of heat explosion, oscillating heat explosion, is discovered. In this case the temperature oscillates during some time remaining bounded, and after that increases exponentially leading to explosion.

In the square domain one or two convective vortices are observed. If we increase the length L_x of the domain, the number of vortices can also increase. They can interact with each other resulting in appearance of complex oscillations including quasi-periodic structurally unstable regimes [2]. Oscillating heat explosion can occur in this case not from periodic oscillation, as in the case of a square domain, but from chaotic oscillations.

As it is indicated above, the boundary condition for the temperature (1.7) provides existence of a stationary solution without convection. If we put $\theta = 0$ at the whole boundary, then the situation is different: there are no regimes without convection even for small R. The behavior of convective solutions is quite different in this case. Complex oscillations occur in a square domain, and another route to chaos, through period doubling bifurcations is observed. Moreover, there are periodic oscillations beyond chaotic with periods corresponding to Sharkovskii sequences [2].

Thus, the boundary condition for the temperature can essentially influence the peculiarities of the process. In this work we will consider heat exchange at the boundary where the heat flux at the boundary is proportional to the temperature, $\partial\theta/\partial n = -\sigma\theta$. We will see that it can change the behavior of oscillating regimes and the transition to explosion.

The contents of the paper are as follows. In the next section we study analytically the one-dimensional model without convection. In Sec. 3 we present numerical results. We introduce a simplified model problem and discuss the results in Sec. 4.

2. Analytical Solution of the 1D Model

In this section we consider the one-dimensional equation

$$\theta'' + ke^{\theta} = 0 \tag{2.1}$$

in the interval $0 \le x \le 2L$ together with the boundary conditions

$$x = 0 : \theta' = \sigma\theta, \qquad x = 2L : \theta' = -\sigma\theta. \tag{2.2}$$

We will find conditions of existence of solutions of this problem, that is critical conditions of heat explosion.

Since the solution, if it exists, is symmetric with respect to $x = L$, we will consider equation (2.1) in the interval $0 \le x \le L$ with the boundary conditions

$$x = 0 : \theta' = \sigma\theta, \qquad x = L : \theta' = 0. \tag{2.3}$$

We will denote by θ_0 the value of the dimensionless temperature at $x = 0$, and by θ_m the maximal temperature reached at $x = L$.

Multiplying (2.1) by θ' and integrating, we obtain

$$(\theta')^2 + 2ke^{\theta} = \sigma^2\theta_0^2 + 2ke^{\theta_0}. \tag{2.4}$$

Taken for $x = L$, this equation gives the relation between θ_0 and θ_m:

$$e^{\theta_m} = e^{\theta_0} + \alpha\theta_0^2, \tag{2.5}$$

where $\alpha = \sigma^2/(2k)$. On the other hand, we can solve (2.4) to obtain the implicit formula for the temperature distribution:

$$\int_{\theta_0}^{\theta(x)} \frac{ds}{\sqrt{2k(e^{\theta_m} - e^s)}} = x. \tag{2.6}$$

Substituting here $x = L$ and taking into account (2.5), we obtain the equation

$$\frac{1}{\sqrt{e^{\theta_0} + \alpha\theta_0^2}} \operatorname{argtanh} \sqrt{\frac{\alpha\theta_0^2}{e^{\theta_0} + \alpha\theta_0^2}} = \sqrt{\frac{k}{2}}\, L \tag{2.7}$$

with respect to θ_0. Here argtanh denotes the argument of the hyperbolic tangent. Denote the left-hand side of this equation by $F(\theta_0)$. The number of solutions of problem (2.1)–(2.2) is related to the number of solutions of Eq. (2.7). Let now $M > 0$ be given. We are interested in the equation

$$F(\theta) = M. \tag{2.8}$$

For that purpose we introduce the function g defined by

$$g(\theta) = \operatorname{argtanh}\sqrt{\frac{\alpha\theta^2}{e^\theta + \alpha\theta^2}} - M\sqrt{e^\theta + \alpha\theta^2}. \tag{2.9}$$

The function g satisfies

$$g(0) = -M < 0 \quad \text{and} \quad g(+\infty) = -\infty. \tag{2.10}$$

Next simple computations show that the equation $g'(\theta) = 0$ is equivalent to

$$\frac{M}{\sqrt{\alpha}} e^\theta = 2 - (1 + 2M\sqrt{\alpha})\theta. \tag{2.11}$$

If $M > 2\sqrt{\alpha}$ then Eq. (2.11) does not have any solution. In that case the function g is decreasing and Eq. (2.8) does not have solution. If $M \le 2\sqrt{\alpha}$ then Eq. (2.11) has only one solution θ^*; the function g is increasing for $0 \le \theta \le \theta^*$ and decreasing for $\theta \ge \theta^*$. Finally, the number of solutions for Eq. (2.8) is related to the sign of the quantity $g(\theta^*)$. Indeed (2.8) does not have solution when $g(\theta^*) < 0$, has only one solution when $g(\theta^*) = 0$ and exactly two solutions when it is non-negative. Thus for k and L sufficiently large, Eq. (2.7) does not have solutions. Therefore, problem (2.1), (2.2) has also no solutions. This situation corresponds to heat explosion.

We are now interested in the limit $\sigma \to +\infty$ which formally corresponds to homogeneous Dirichlet conditions $\theta(0) = \theta(2L) = 0$.

Recalling the definition of $\alpha = \sigma^2/(2k)$, we obtain that for sufficiently large σ, equation (2.11) has one solution θ^*_σ. The asymptotic analysis for large σ shows that

$$\theta^*_\sigma = \frac{\sqrt{2k}}{M\sigma} - \left(1 + \frac{1}{M^2}\right)\frac{k}{\sigma^2} + o\left(\frac{1}{\sigma^2}\right).$$

Then we obtain that

$$g(\theta^*_\sigma) = \operatorname{argtanh}\sqrt{\frac{M^2}{1 + M^2}} - \sqrt{1 + M^2} + O\left(\frac{1}{\sigma}\right). \tag{2.12}$$

We observe that leading term in the right-hand side of (2.12) is increasing for small values $M \le M^*$ and decreasing for $M \ge M^*$. It also tends to $-\infty$ for large values of M. For large σ, the critical condition of explosion corresponds to the following equation

$$\operatorname{argtanh}\sqrt{\frac{M^2}{1 + M^2}} = \sqrt{1 + M^2}. \tag{2.13}$$

Finally, we can apply this asymptotics to Eq. (2.7), which corresponds to (2.8) with $M = \sqrt{\frac{k}{2}}L$. It can be verified that the critical value of k obtained above converges to the critical value $k_c \approx 0.88$ for $L = 1$ obtained for the case of the zero boundary conditions [1].

3. Numerical Simulations

In this section we present results of the numerical simulations of the complete problem. We consider the Navier–Stokes equations in the stream function – vorticity formulation:

$$\frac{\partial\theta}{\partial t} + u\frac{\partial\theta}{\partial x} + v\frac{\partial\theta}{\partial y} = \Delta\theta + ke^{\theta}, \tag{3.1}$$

$$\frac{\partial\omega}{\partial t} + u\frac{\partial\omega}{\partial x} + v\frac{\partial\omega}{\partial y} = -PR\frac{\partial\theta}{\partial x} + P\Delta\omega, \tag{3.2}$$

$$-\Delta\Psi = \omega. \tag{3.3}$$

Here ω is the vorticity and Ψ is the stream function

$$u = \frac{\partial\Psi}{\partial y}, \qquad v = -\frac{\partial\Psi}{\partial x}. \tag{3.4}$$

Equations (3.1)–(3.3) are completed by the boundary conditions

$$\frac{\partial\theta}{\partial n} + \sigma\theta = 0, \tag{3.5}$$

$$\omega = 0, \qquad \Psi = 0. \tag{3.6}$$

This boundary condition corresponds to the free surface boundary conditions for the velocity.

We use finite difference approximations and the alternating directions method to solve the problem numerically.

The results of the numerical simulations are summarized in Fig. 1. On the parameter plane (R, k) and for fixed values of other parameters $L_x = 8,\ L_y = 2,\ \sigma = 50,\ P = 3$, we show the region with stable stationary convective regimes (below the lower curve) and the region where explosion occurs (above the upper curve). There is also the third region (between two curves) where the temperature remains bounded but the stationary solution is not stable. Instead of it we observe temporal oscillations that can be periodic or aperiodic.

It is interesting to note that oscillations appear only for R sufficiently large. For small R there is a direct transition from a stable stationary convective solution to explosion where the temperature increases and becomes unbounded.

We present a series of simulations for $R = 5000$ and for different values of k. A stationary temperature distribution and the stream function for a value of k just below the lower curve ($k = 0.93$) are shown in Fig. 2. There are two vortices symmetric with respect to the central line of the domain.

For k just above the lower curve ($k = 0.931$) there are periodic oscillations. Figure 3 shows the temperature and the stream function in consecutive moments of time. The structure of the oscillations is shown in Fig. 4. The maximum of the stream function (absolute value) is a periodic function of time (left figure). During approximately half-period it remains constant, the level lines of the stream function (Fig. 3) show a well

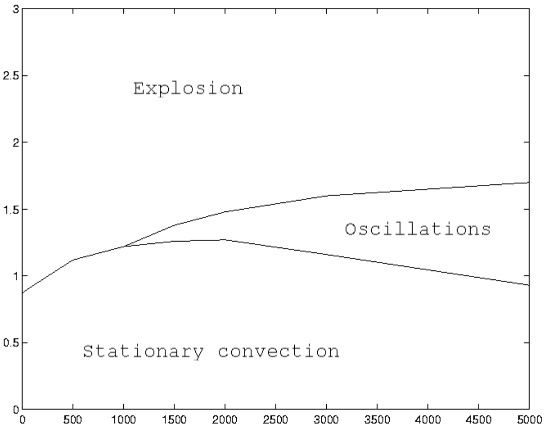

FIG. 1. Behavior of solutions in the parameter plane (R, k).

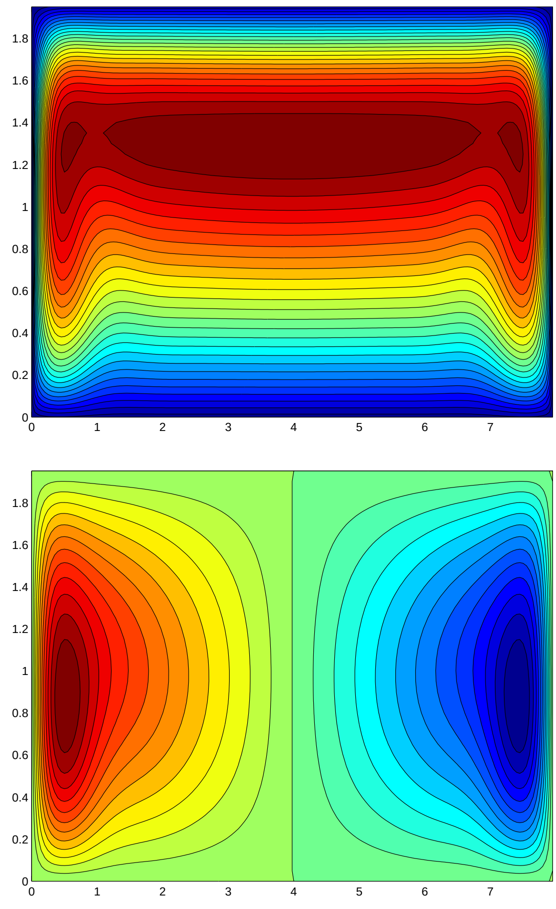

FIG. 2. Stationary distribution of the temperature on the top and level lines of the stream function on the bottom for $k = 0.93$.

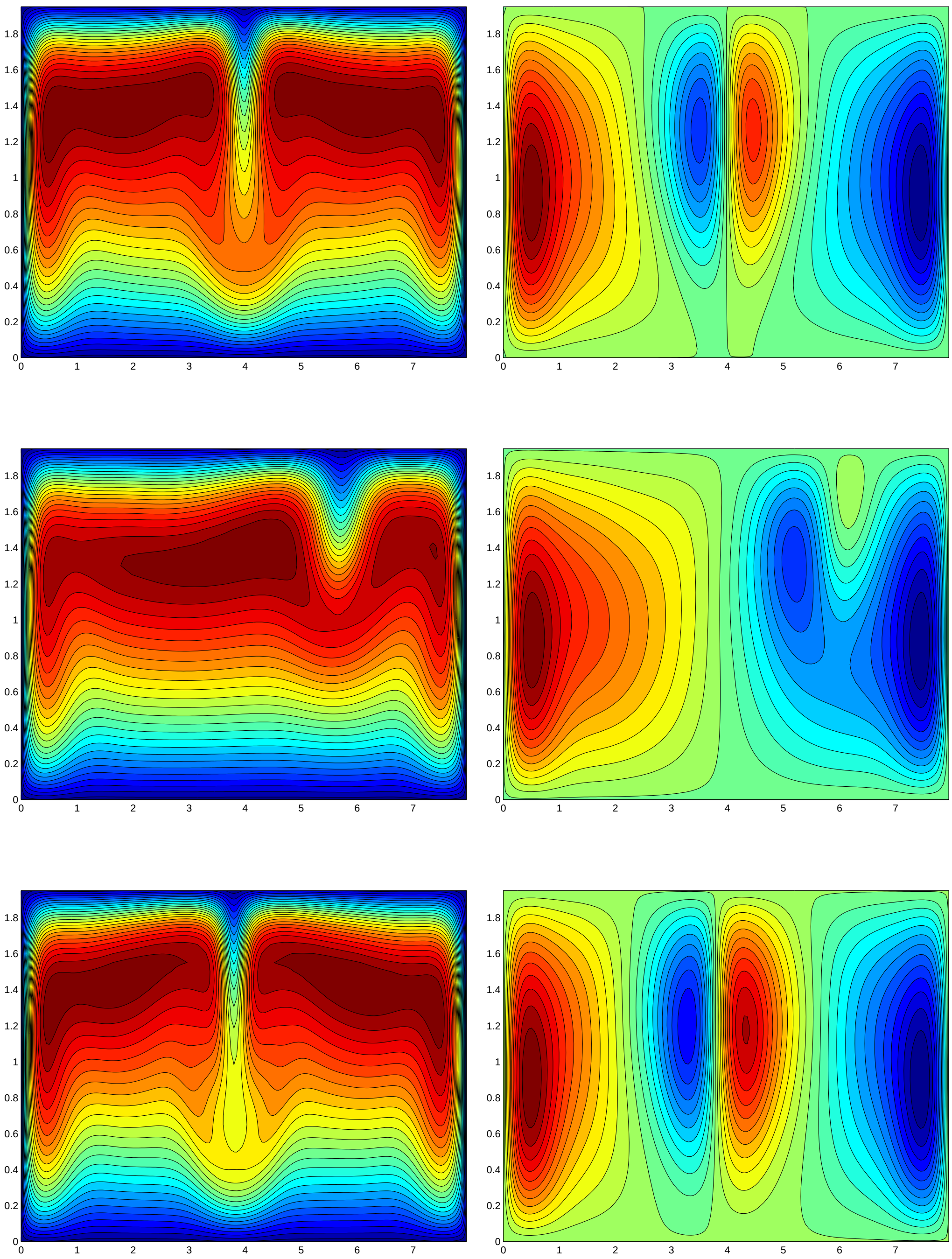

Fig. 3. Temperature distribution (left) and level lines of the stream function (right) for consecutive moments of times for $k = 0.931$.

ordered structure with four vortices. During the second half-period the stream function rapidly oscillates, and the structure of convective rolls changes. Figure 4 (right) represent the same results at the (θ, ψ)-plane, where θ is the mean value of the temperature at each given moment of time, and ψ is the mean of the stream function. Since the solution is periodic, the "trajectory" is closed but it has a complex structure with a number of "branches" above and below the line $\psi = 0$. In fact, the curve is symetric with respect to this line. This means that the stream function changes its sign, that is the structure of the vortices is the same but they rotate in the opposite directions.

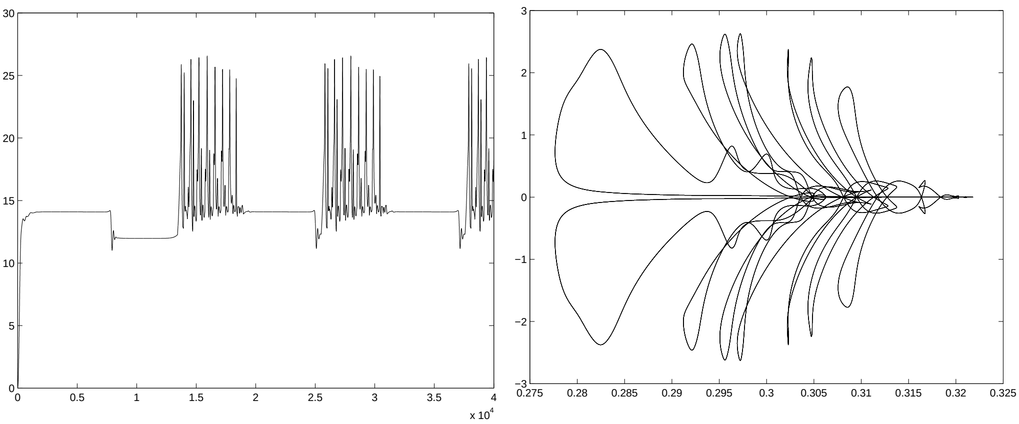

FIG. 4. Evolution of the maximum of the stream function (left) for $k = 0.931$, phase plane $(\theta - \psi)$ (right).

Increasing k we observe a period doubling sequence of bifurcations. For $k = 0.932$ we obtain a 4-perodic trajectory (see Fig. 5) and for $k = 0.93205$ we obtain a 7-periodic curve Fig. 6. By analogy with the Sharkovskii sequences, there are chaotic oscillations for k between these two values.

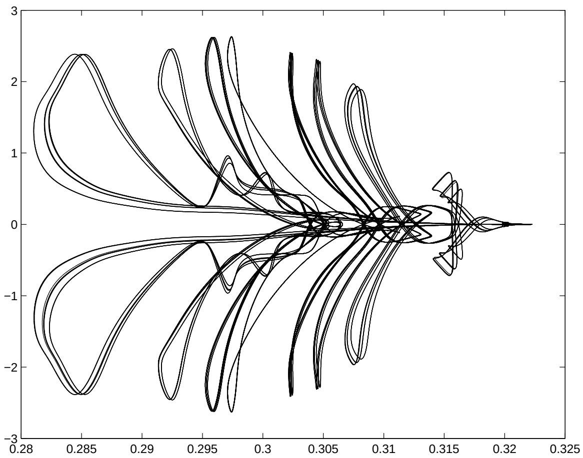

FIG. 5. 4-periodic trajectory in the phase plane $(\theta - \psi)$ for $k = 0.932$.

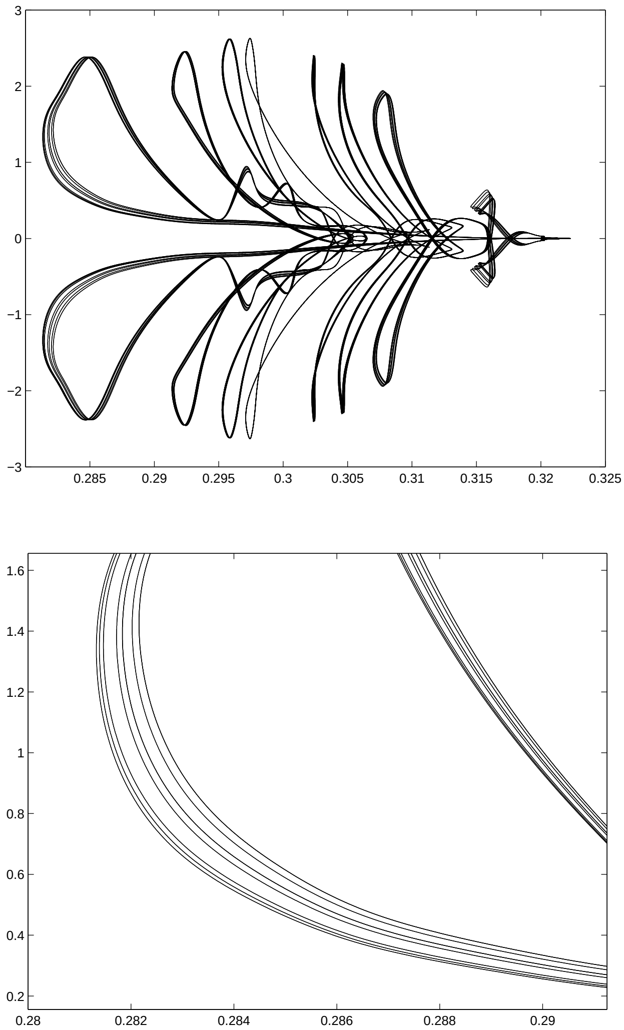

FIG. 6. 7-periodic trajectory in the phase plane $(\theta - \psi)$ for $k = 0.93205$.

For a larger value of k ($k = 0.958$) there are periodic oscillations with a simpler structure (Figs. 7, 8). Further increase of this parameter ($k = 0.9999$) leads again to a period doubling bifurcation (Fig. 9).

Further increase the parameter k leads to appearance of an aperiodic trajectory (Fig. 10). For k sufficiently large the temperature increases resulting in heat explosion. Figure 11 shows the maximal temperature as a function of time for k, right above the upper curve. It is interesting to note that the temperature begins to oscillate at the same time as it grows. Such modes of explosion were not observed before. We suppose that this is related to the type of boundary conditions considered in this work. We will discuss this question in the next section.

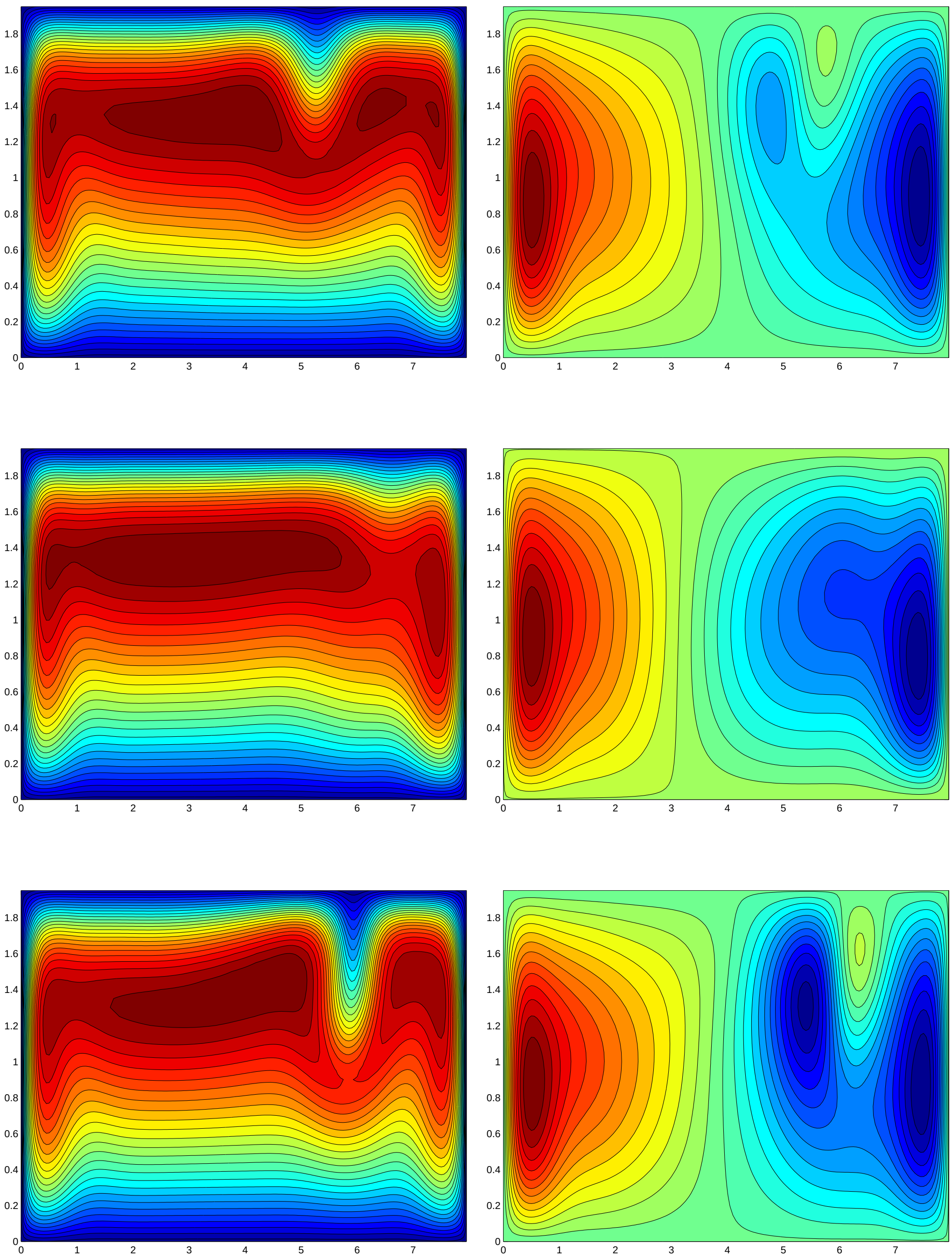

FIG. 7. Temperature distribution (left) and level lines of the stream function (right) for consecutive moments of times for $k = 0.958$.

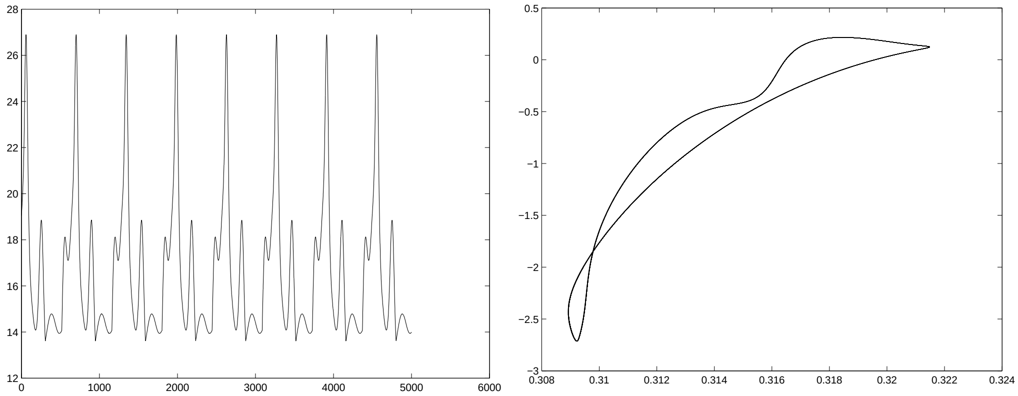

Fig. 8. Evolution of the maximum of the stream function (left), phase plane (right) for $k = 0.958$.

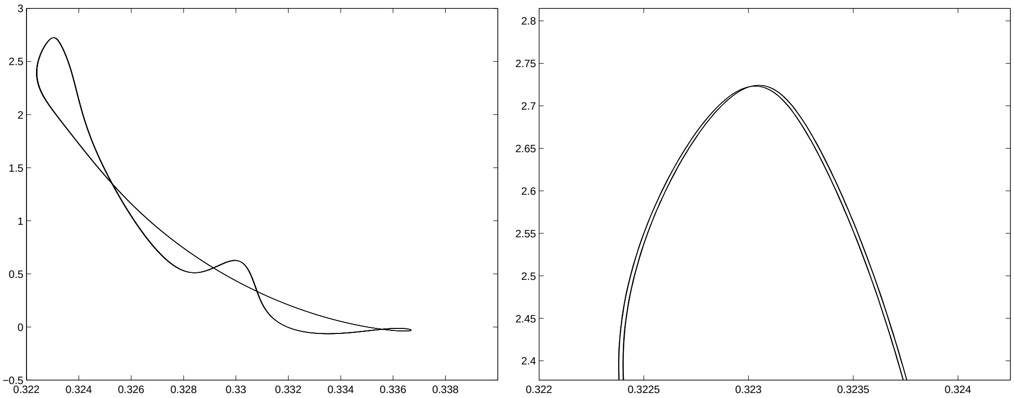

Fig. 9. 2-periodic trajectory in the phase plane for $k = 0.9999$.

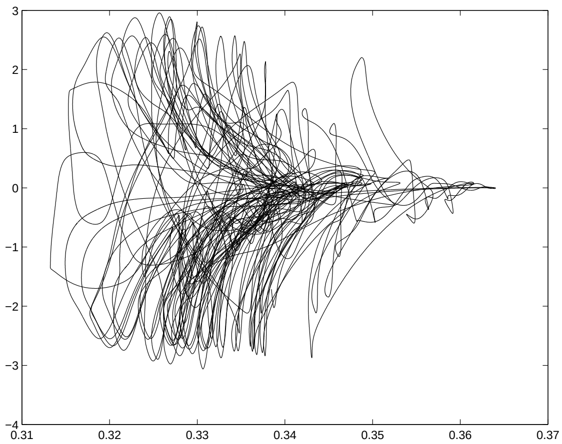

Fig. 10. Aperiodic trajectory in the phase plane for $k = 1.02$.

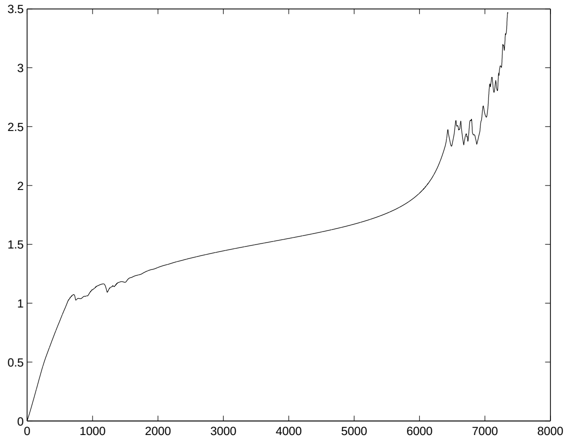

FIG. 11. Oscillating explosion corresponding to $k = 1.72$ and $R = 5000$.

4. Simplified Model Problem

In Semenov's model of heat explosion the coefficient of heat loss α is a constant. In the presence of convection the intensity of heat loss depends on the intensity of convection that can be measured by an average value ψ of the stream function. Therefore we should consider α as a function of ψ. To have a closed system we need then to introduce an equation for ψ. Numerical simulations of the complete problem shows that for stationary solutions the average value of the stream function can be well approximated as a linear function of an average temperature [2]. Therefore we obtain the system of two ordinary differential equations

$$\frac{d\theta}{dt} = K(\theta) - \alpha(\psi)\theta, \qquad \frac{d\psi}{dt} = a\theta - b\psi, \tag{4.1}$$

where a and b are positive parameters, $\alpha(\psi)$ can be considered for simplicity as a linear function. It is a generalization of Semenov's model of heat explosion for the case with convection. Though the model is very simple, it shows the main features of the complete model including periodic oscillations and oscillating heat explosion [2].

This model is analyzed in [2]. We briefly recall here its main properties. In the most interesting for us case, this system has two stationary points, one of them is a saddle, and another one a stable or unstable focus. The key feature of this system is that it can have a homoclinic orbit starting from the saddle and coming back to it, and containing the second point inside it. As it is well known, stable or unstable periodic orbits can bifurcate from a homoclinic orbit. The oscillating heat explosion corresponds to the bifurcation of an unstable periodic orbit. In this case the solution will oscillate during some time remaining close to the periodic orbit, and after some time it will leave it and go to infinity. This behavior was observed for the complete and for the simplified models.

Thus we can explain, in the framework of this model, appearance of oscillations before the final temperature growth in the process of explosion. However we cannot explain the oscillations described above that happen at the same time with the temperature growth. For this we need to modify the model system taking into account another type of boundary conditions for the temperature.

Along with the average temperature θ inside the domain, let us also consider the average temperature θ_b at the boundary. The difference between them is proportional to the average value of the temperature gradient. Therefore, at the same time the heat loss and the intensity of convection will be proportional to $\theta - \theta_b$. Instead of system (4.1) we consider in this case

$$\frac{d\theta}{dt} = K(\theta) - \alpha(\psi)(\theta - \theta_b), \qquad \frac{d\psi}{dt} = a(\theta - \theta_b) - b\psi. \tag{4.2}$$

It is a phenomenological model justified by the physical arguments presented above.

An important property of this system is that it is not closed. The value θ_b is not known. It changes in time, and it cannot be found as a solution of (4.2). We will consider it as a given and increasing function of time. Moreover, the parameter b in the second equation of (4.2) should be also considered now as a function of time. Indeed, we can find from this equation the stationary value of ψ:

$$\psi = \frac{a(\theta - \theta_b)}{b}.$$

If θ_b grows but the difference $(\theta - \theta_b)$ remains constant, then according to this formula, the intensity of convection will not change either. However it is different for the complete problem where the increase of the temperature at the boundary leads to the growth of the temperature gradient at the boundary, which reinforces convection. To take into account this effect in the simplified model, we should consider b as a decreasing function of time.

Suppose next that the dependences of θ_b and b on t are weak. Then we can apply a quasi-stationary approximation and study system (4.2) as it is done in the case of constant coefficients. As before, it can have two stationary points and an unstable periodic orbit. The difference with respect to the previous case is now that the stationary points and the periodic orbit move in time in the direction of higher temperatures. Thus we can obtain temperature oscillations at the same time with the temperature growth in the process of explosion.

References

1. J. Bebernes, D. Eberly, *Mathematical problems from combustion theory*, Applied Mathematical Sciences, 83, Springer Verlag, New York 1989.

2. M. Belk, V. Volpert, *Modelling of heat explosion with convection*, Chaos, **14**, 263–279, 2004.

3. T. Dumont, S. Génieys, M. Massot, V. Volpert, *Interaction of thermal explosion and natural convection: critical conditions and new oscillating regimes*, Siam J. Appl. Math., **63**, 351–372, 2002.

4. D.A. Frank-Kamanetskii, *Diffusion and heat transfer in chemical kinetics*, Plenum, New York 1969.

5. A.S. Merzhanov, E.A. Shtessel, *Free convection and thermal explosion in reactive systems*, Astronautica Acta, **18**, 191–199, 1973.

6. N.N. Semenov, *To the theory of combustion processes*, Zh. Fiz. Khim., **4**, 4–17, 1933.

7. N.N. Semenov, *Thermal theory of combustion and explosions*, Usp. Fiz. Nauk, **23**, 4–17, 1940.

8. Y.B. Zeldovich, G.I. Barenblatt, V.B. Librovich, G.M. Makhviladze, *The mathematical theory of combustion and explosions*, Plenum, New York 1985.

Received November 9, 2004.