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HYDRODYNAMIC MECHANISM OF SPONTANEOUS RETARDATION OF AN ENDOTHERMIC REACTION

V. I. Naidenov

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It is known that the steady operating regimes of continuous reactors for endothermic reactions are generally stable. This is true if the velocity of the reacting mixture is not dependent on temperature and concentration. However, as was shown in [2-4], allowing for the dependence of the viscosity of the fluid and, thus, the flow velocity on the governing factors of the process (temperature, degree of conversion) leads to qualitative changes in the processes of chemical conversion, heat and mass transfer, and motion. The studies [5-7] examined nonlinear effects in media in the case of volumetric heat adsorption, these effects having been due to the temperature dependence of the thermal conductivity and the capacity of the heat sink.

Here we describe the hydrodynamic mechanism of spontaneous retardation of an endothermic reaction occurring in a moving homogeneous fluid. It should be noted that the results obtained are also valid for other manufacturing processes which involve the motion of a fluid with variable viscosity.

We will examine the laminar flow of a viscous fluid in a circular tube of radius a and length L under the influence of an applied pressure gradient p. For simplicity, we will assume that there is no heat transfer through the tube wall and that an endothermic reaction with an activation energy E_2 and a thermal effect q is occurring in the fluid. We further assume that the absolute viscosity of the fluid is determined by the Arrhenius dependence on absolute temperature

$$\mu = \mu_0 \exp \frac{E_1}{R}\left(\frac{1}{T} - \frac{1}{T_0}\right),$$

where μ_0 and T_0 are the viscosity and temperature of the fluid at the inlet; E_1, activation energy for viscous flow; R, gas constant.

Assuming the Reynolds number to be small ($\mathrm{Re} = Q\rho/\pi a\mu_0$), we use a hydraulic approximation of the equations of motion and heat transfer

$$p = 8\mu_0 e^{\frac{E_1}{R}\left(\frac{1}{T}-\frac{1}{T_0}\right)} \frac{Q}{\pi a^4}, \tag{1}$$

$$\frac{dT}{dt} + \frac{Q}{\pi a^2 L\rho c_p}(T - T_0) = -\frac{q}{\rho c_p} e^{-\frac{E_2}{RT}} + \frac{\mu_0 Q^2}{\pi^2 a^6 \rho c_p} e^{\frac{E_1}{R}\left(\frac{1}{T}-\frac{1}{T_0}\right)},$$

where Q is the fluid flow rate; ρ and c_p are the density and specific heat of the fluid. System (1) in essence describes the processes of motion and heat transfer in an ideal-

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mixing endothermic reactor [1].

We introduce dimensionless quantities and write system (1) in the form

$$\omega = e^{-1/\theta},\quad \frac{d\theta}{d\tau} + \gamma\omega(\theta - \theta_0) = - e^{-\beta/\theta} + \mathrm{Br}\,\omega^2 e^{1/\theta}, \tag{2}$$

$$\theta = \frac{RT}{E_1},\quad \omega = \frac{8\mu_0 Q}{\pi a^4 p} e^{-1/\theta_0},\quad \tau = \frac{qR}{\rho c_p E_1} t,$$

$$\theta_0 = \frac{RT_0}{E_1},\quad \gamma = \frac{a^2 E_1}{8\mu_0 q L R} e^{1/\theta_0} p,\quad \beta = \frac{E_2}{E_1},$$

$$\alpha = \frac{\mu_0 q L^2 R^2}{a^2 E_1} e^{-1/\theta_0},\quad \mathrm{Br} = \alpha\gamma^2.$$

Here, the parameters γ and Br respectively characterize the ratio of the amount of heat transferred by convection and liberated in the fluid as a result of frictional forces to the amount of heat absorbed in the chemical reaction; the parameter β determines the ratio of the rates of heat liberated and heat absorbed.

We will study the state of thermal equilibrium θ_s, ω_s:

$$\omega_s = e^{-1/\theta_s},$$
$$\gamma\omega_s(\theta_s - \theta_0) = - e^{-\beta/\theta_s} + \mathrm{Br}\,\omega_s^2 e^{1/\theta_s}. \tag{3}$$

Excluding the fluid velocity ω_s, we have the following equation to determine the temperature θ_s

$$\gamma(\theta_0 - \theta_s) + \mathrm{Br} = e^{(1-\beta)/\theta_s}. \tag{4}$$

We will examine the case $\beta < 1$ (the activation energy for viscous flow exceeds the activation energy for the chemical reaction). Analyzing (4), we obtain γ^* such that at $\gamma < \gamma^*$ there are no steady-state solutions to system (2). At $\gamma = \gamma^*$, there is a unique state of thermal equilibrium. At $\gamma > \gamma^*$, there is a singularity. Each value of $\gamma \in (\gamma^*, \infty)$ corresponds to two values of temperature $\theta_s^{1,2}$ and fluid velocity $\omega_s^{1,2}$. To determine the critical values θ_s^* and ω_s^*, we find from Eq. (4) that

$$\frac{d\gamma}{d\theta_s}(\theta_s^*) = 0, \tag{5}$$

$$\theta_s^{*2}\left[\theta_s^{*2} - (\theta_0 - \theta_s^*)(1-\beta)\right] = \alpha(1-\beta)^2 e^{(1-\beta)/\theta_s^*},$$

$$\gamma^* = (1-\beta)\,\theta_s^{*-2} e^{(1-\beta)/\theta_s^*}.$$

We will study the stability of steady-state solutions θ_s and ω_s at $\gamma > \gamma^*$. We seek nonsteady solutions of system (2) in the form $\theta = \theta_s + \theta'$, $\omega = \omega_s + \omega'$. Inserting these functions into (2) and linearizing (2) in the neighborhood of the equilibrium position, we write the equation for the temperature perturbation

$$\frac{d\theta'}{d\tau} = \frac{e^{-1/\theta_s}\theta'}{\theta_s^2}\left[(1-\beta)e^{(1-\beta)/\theta_s} - \gamma\vartheta_s^2\right].$$

The solution of this equation has the form $\theta' = e^{\lambda\tau}$, and to determine λ we obtain the spectral equation

$$\lambda = e^{-1/\theta_s}\theta_s^{-2}\left[(1-\beta)e^{(1-\beta)/\theta_s} - \gamma\theta_s^2\right].$$

Simple analysis shows that at $\beta < 1$ $\theta_s^1 < \theta_s^*$, $\omega_s^1 < \omega_s^*$, $\lambda > 0$; $\theta_s^2 > \theta_s^*$, $\omega_s^2 > \omega_s^*$, $\lambda < 0$, i.e., the regimes of heat transfer and motion with low temperatures and velocities turn out to be unstable in regard to infinitely small thermal perturbations. This should be expected, since the low-temperature state of equilibrium is characterized by the relation $dp/dQ < 0$ (the mass flow of the fluid decreases with an increase in the pressure gradient), and such regimes are usually energetically unstable [8]. At $\beta > 1$ and $\lambda < 0$, there is a unique stable state of thermal equilibrium.

Let us examine the nonsteady problem

$$d\theta/d\tau = -e^{-\beta/\theta} - \gamma e^{-1/\theta}(\theta - \theta_0) + \mathrm{Br}\, e^{-1/\theta}. \qquad (6)$$

Analyzing (6), we find at $\beta = 0$ $d\theta/d\tau \to -1$ for $\theta \to 0$, while at $\beta > 0$ $d\theta/d\tau \to 0$ for $\theta \to 0$, i.e., the rate of cooling of the fluid after loss of stability is greater, the greater the difference in the activation energies for viscous flow and the chemical reaction. The regions of the critical temperatures $\theta_s^* > \theta_0$ obviously correspond to the region of substantial dissipative heat liberation; in the region $\theta_s^* < \theta_0$, heat convection predominates. With an increase in θ_s^*, the critical pressure gradient decreases, since there is a decrease in the viscosity of the fluid.

Let us turn to physical analysis of the results obtained. We will study the region of interaction of heat convection and absorption (the dissipation of mechanical energy is small compared to these factors of the process). This region corresponds to critical temperatures of the fluid less than the inlet temperature. In the case of large pressure gradients or small thermal effects in the reaction, the flow of heated viscous fluid introduces a sufficient quantity of heat for the reaction to proceed in the steady-state regime. In this case, two equilibrium states are established due to the presence of nonlinear relationships in the system. One of these states, with a higher fluid velocity and temperature, is stable. The other equlibrium state is unstable in regard to small thermal perturbations. In the case of small pressure gradients or large thermal effects in the reaction, a situation is created whereby the amount of heat introduced by the fluid flow is insufficient to ensure steady-state conditions of motion and heat transfer. The structure of the heat balance undergoes a qualitative change and since heat absorption will be greater than heat liberation, a nonsteady regime of motion of the viscous fluid develops, and the endothermic reaction takes place with a progressive decrease in temperature, fluid velocity, and reaction rate.

The critical flow regime dividing qualitatively different types of fluid motion (steady and nonsteady) is unstable in regard to thermal perturbations; if the temperature of the system is lowered as a result of random perturbations, viscosity will increase and convective heat transfer will decrease, temperature will decrease further, etc. Naturally, the rate of heat absorption will also decrease in this case. However, since the activation energy for viscous flow is greater than the activation energy for the endothermic reaction, the decrease in the capacity of the heat sink will take place more slowly than the decrease in thermal convection. The increase in the temperature gradient along the reaction zone – a stabilizing factor in the situation being examined – will not play a significant role because the total rate of heat liberation and heat absorption depends exponentially on temperature and suppresses its change along the flow.

The physical phenomenon of a decrease in the rate of heat absorption resulting from a progressive increase in the viscosity of the fluid medium can be regarded as spontaneous hydrodynamic retardation of the endothermic reaction. If the activation energy for viscous flow is significantly greater than the activation energy of the chemical reaction, then spontaneous retardation will take place more abruptly because there is a definite time interval over which fluid velocity reaches very high values. Here, the drop in fluid temperature is catastrophic in character and occurs over a relatively short period of time which can be regarded as the induction period of the phenomenon under discussion.

Where the activation energies for viscous flow and the reaction approach each other, a decrease in the temperature and an increase in the viscosity of the fluid will take place over a long time interval and be asymptotic in character. The process of spontaneous hydrodynamic retardation of heat absorption will be progressive output of energy to the external source maintaining the motion of the fluid. Such a spontaneous physical cessation of the chemical reaction, due to the hydrodynamic conditions of its occurrence, may be of interest for a certain range of phenomena involving spontaneous organization in nonequilibrium systems [9]. Due to the presence of positive feedback in the strongly nonequilibrium thermodynamic system being examined, the chemical reaction, controlling the delivery of the necessary energy, may cut off its supply by slowing the flow of fluid carrying heat.

The mechanism of spontaneous retardation is somewhat different in the region of substantial dissipative heat liberation. The heat given off in the fluid due to internal friction is absorbed by the reaction and removed through the cross section of the tube by convection. Since the rate of heat liberation is roughly the same as the rate of convective heat transfer (both processes are determined by the fluid velocity), then the mechanism by which thermal instability develops is in principle the same as before: beginning with a certain critical pressure gradient, the heat source ceases to balance the sink. If fluid velocity is less dependent on temperature than the rate of heat absorption, then critical phenomena are not observed; in this case, a decrease in fluid temperature leads to an equivalent decrease in the capacity of the heat sink, thereby stabilizing the processes of heat transfer and motion.

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MODEL OF THE TAYLOR INSTABILITY

M. D. Kamchibekov

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1. The Taylor (or Rayleigh–Taylor) instability arises on the boundary between a heavy and a light liquid in the field of gravity, when the heavy liquid is above the light one. The Taylor instability has been studied actively both experimentally [1] and theoretically with analytical methods [2-5], and by numerical simulation of the development of the instability [6-8]. Interest in this phenomenon has increased in recent years in connection with problems of control of thermonuclear synthesis with inertial confinement of a plasma. It was shown in [9, 10] that the development of an instability in the surface layers of a target is a Taylor instability and can lead to the deterioration of the symmetry of compression and characteristics of the target. In calculating the development of the Taylor instability in such problems a simple model is useful, which should describe the growth of perturbations with acceptable accuracy for practical purposes. In the present paper, using physically justifiable assumptions about the form of the perturbation, we simplify the formation of the problem on the development of the Taylor instability and obtain equations which correctly describe both the linear and asymptotic stages of the Taylor instability.

2. The usual formulation of the Taylor instability problem is as follows [4]. In a uniform gravity field with acceleration g directed downward, a heavy liquid with density ρ occupies the space $z > \theta(x, t)$ and a light liquid with density $\rho_*(\rho_* < \rho)$ occupies the space $z < \theta(x, t)$, where $\theta(x, t)$ is the vertical position of the surface between the heavy and light liquids, and z and x are the vertical and horizontal Cartesian coordinates. At the time t = 0, the initial perturbation of the dividing surface is specified as

$$\theta(x,\ t=0)=\theta_0\cos kx \tag{2.1}$$

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