



Influence of vibrations on convective instability of polymerization fronts

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Abstract. Propagation of polymerization fronts with liquid monomer and solid polymer is considered, and the influence of vibrations on critical conditions of convective instability is studied. The model includes the heat equation, the equation for the concentration and the Navier-Stokes equations considered under the Boussinesq approximation. Linear stability analysis of the problem is fulfilled, and the convective instability boundary is found depending on the amplitude and on the frequency of vibrations.

Key words: convective instability, frontal polymerization, vibrations.

1. Introduction

There are various types of instabilities of reaction fronts, including thermo-diffusional instability, the hydrodynamical instability, and the convective instability.

The thermo-diffusional instability appears as a result of competition between the heat production in the reaction zone and heat transfer to the cold reactants. To study this type of instability, the density of the medium can be taken constant to remove the influence of hydrodynamics and to simplify the model. The stability conditions in this case were studied in [1–5].

A hydrodynamic instability, also called Darrieus-Landau instability, of the reaction front can occur if the density of the medium is variable. Usually it is considered as a given function of the temperature. The instability is caused by heat expansion of the gas or liquid in a neighbourhood of the reaction zone [6–10].

By convective instability, we mean that one which appears due to natural convection. This instability can be studied in the pure form if we consider the Boussinesq approximation, *i.e.*, we neglect the change of density everywhere except for the buoyancy term. The Boussinesq approximation was justified and used to study the front stability in [4, 11].

It is known that vibrations can influence conditions of the hydrodynamic instability of gaseous flames [19, 20], convective instability of directional solidification [17], [26] and crystall growth [27–29]. In this work we study the influence of vibrations on convective instability of polymerization fronts.

Convective instability of polymerization fronts without vibrations was studied in [12–16]. A typical example of polymerization front is shown in Figure 1. In this case the monomer is liquid, the polymer is solid, and they are separated by a narrow reaction zone. This is a thermal front, which propagates if the reaction is highly activated and exothermic. The exother-

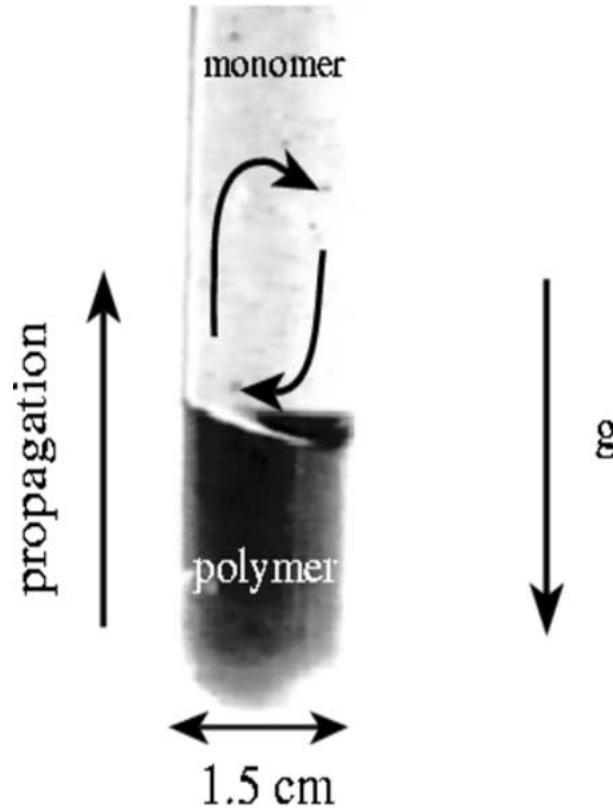


Figure 1. Propagation of a polymerization front with convection. The system consists of acrylamide dissolved in dimethyl sulfoxide with 5% bis-acrylamide as a crosslinking agent and sodium persulfate as the initiator. Adapted from [14].

mic reaction heats the liquid monomer from below. The critical conditions of appearance of convection are determined by the frontal Rayleigh number,

$$R = \frac{g\beta q\kappa^2}{\nu c^3},$$

where g is the gravity acceleration, β the coefficient of thermal expansion, q the adiabatic heat release, κ the coefficient of thermal conductivity, ν the kinematic viscosity, and c is a speed of propagation of the front.

If the frontal Rayleigh number exceeds a critical value, then convection appears. Depending on values of the parameters, two different convective structures are observed [12–14]. One of them is antisymmetric with one vortex (Figure 1), another one is axisymmetric with two vortices.

This work is devoted to the influence of vibration on polymerization fronts. Vibrations are known to be among the most effective ways to affect the behaviour of a fluid. The periodic oscillations of the containing vessel may influence the convective instability. In order to study the influence of vibrations on the instability of a polymerization front, we impose a harmonic oscillation of frequency σ , and amplitude λ in the vertical direction upon the system containing the liquid monomer and the solid polymer. This oscillation causes a periodic acceleration, b , perpendicular to the liquid-solid interface. The time dependence of the instantaneous accel-

eration acting on the fluids is then given by $g + b(t)$, where $b(t) = \lambda \sin(\sigma t)$. In this work we use the Boussinesq approximation, which takes into account the temperature dependence of the density only in the volumetric forces.

This study is motivated by two types of applications. The first one is related to polymer technologies: continuous polymerization reactors [22, 23] and reactive injection molding (RIM) [24, 25]. Frontal polymerization is a simplified model of these processes where propagation of a localized reaction zone can be studied independently of reactive flows. Vibrations can be used to suppress the convective instability or, inversely, to enforce it when we need to mix the reactants.

This is a first step in studying vibration effects on polymerization fronts. Here we consider the case with a solid product, which closely relates to RIM while the case with liquid product is similar to adiabatic tubular reactors. We should distinguish between isothermal tubular reactors and adiabatic ones. Some isothermal tubular reactors are over 1 km in length! The former have little relation to frontal polymerization while the later ones are related but they are rare.

The second application is connected with microgravity studies on frontal polymerization [30–32]. Residual acceleration and vibrations, ‘g-jitter’, have been shown to affect many fluid problems [33–36], and we should be able to estimate their impact.

There have been some experimental investigations on the effects of vibration on fluid stability. Wolf studied the stabilization of the Rayleigh-Taylor instability by vibration [37] as did Briskman [38].

Bowden *et al.* studied ascending fronts of free-radical acrylamide polymerization in dimethyl sulfoxide using persulfate as the thermal initiator. By adding ultrafine silica gel they could systematically vary the viscosity [14]. The same system and procedures could be used with the reactor attached to a vibration test stand.

The contents of this work are as follows. We introduce the model in Section 2 and reduce it to a singular perturbation problem in Section 3. The linear stability of the interface problem is fulfilled in Section 4. We discuss the results in Section 5. Conclusions are given in Section 6.

2. Governing equations

We consider the effect of periodic oscillations on propagating polymerization fronts. The process is described by the system of equations:

$$\frac{\partial T}{\partial t} + v \nabla T = \kappa \Delta T + q K(T) \phi(\alpha), \quad (2.1)$$

$$\frac{\partial \alpha}{\partial t} + v \nabla \alpha = K(T) \phi(\alpha), \quad (2.2)$$

$$\frac{\partial v}{\partial t} + (v \nabla) v = -\frac{1}{\rho} \nabla p + v \Delta v + g (1 + \lambda \sin(\sigma t)) \beta (T - T_0) \gamma, \quad (2.3)$$

$$\operatorname{div} v = 0, \quad (2.4)$$

with the conditions at infinity:

$$T = T_i, \quad \alpha = 0 \quad \text{and} \quad v = 0, \quad \text{as } z \rightarrow +\infty,$$

and

$$T = T_b \quad \text{and} \quad v = 0, \quad \text{as } z \rightarrow -\infty.$$

Here T is the temperature, α the depth of conversion, $v = (v_x, v_y, v_z)$ the velocity of the medium, p the pressure, κ the coefficient of thermal diffusivity, q the adiabatic temperature rise, ρ an average value of density, ν the coefficient of kinematic viscosity, g the gravitational acceleration, λ and σ stand for the amplitude and the frequency of vibrations, β the coefficient of thermal expansion, and γ the unit vector in the z -direction (upward),

$$\nabla = \left(\frac{\partial}{\partial x}, \frac{\partial}{\partial y}, \frac{\partial}{\partial z} \right), \quad \Delta = \left(\frac{\partial^2}{\partial x^2}, \frac{\partial^2}{\partial y^2}, \frac{\partial^2}{\partial z^2} \right),$$

x, y, z the spatial coordinates, $-\infty < x, y, z < +\infty$, t time, T_0 the mean value of temperature, T_i is an initial temperature and T_b is the adiabatic temperature, $T_b = T_i + q$, W is the reaction rate,

$$W = K(T)\phi(\alpha).$$

The temperature dependence of the reaction rate is given by the Arrhenius exponential,

$$K(T) = k_0 \exp(-E/R_0T),$$

where E is the activation energy, R_0 the gas constant and k_0 the pre-exponential factor. For the asymptotic analysis of the problem we will assume that the activation energy is large. We consider a zero-order reaction,

$$\phi(\alpha) = \begin{cases} 1 & \text{if } \alpha < 1, \\ 0 & \text{if } \alpha = 1. \end{cases}$$

We introduce dimensionless spatial variables xc_1/κ , yc_1/κ , zc_1/κ , time tc_1^2/κ , velocity v/c_1 and pressure $\frac{p}{(c_1^2\rho)}$. Here $c_1 = c/\sqrt{2}$, c denotes the velocity of the propagation of the stationary front, which can be calculated asymptotically for large Zeldovich number, $Z = \frac{qE}{R_0T_b^2}$ [39]:

$$c^2 = \frac{2k_0\kappa}{q} \frac{R_0T_b^2}{E} \exp(-E/R_0T_b).$$

Denoting $\theta = (T - T_b)/q$ and keeping for convenience the same notation for other variables, we rewrite the system (2.1)–(2.4) in the form:

$$\frac{\partial \theta}{\partial t} + v \nabla \theta = \Delta \theta + Z \exp\left(\frac{\theta}{Z^{-1} + \delta \theta}\right) \phi(\alpha), \quad (2.5)$$

$$\frac{\partial \alpha}{\partial t} + v \nabla \alpha = Z \exp\left(\frac{\theta}{Z^{-1} + \delta \theta}\right) \phi(\alpha), \quad (2.6)$$

$$\frac{\partial v}{\partial t} + (v \nabla) v = -\nabla p + P \Delta v + PR(1 + \lambda \sin(\mu t))(\theta + \theta_0) \gamma, \quad (2.7)$$

$$\operatorname{div} v = 0, \quad (2.8)$$

with the conditions:

$$z \rightarrow +\infty, \quad \theta = -1, \quad \alpha = 0, \quad v = 0, \quad (2.9)$$

$$z \rightarrow -\infty, \quad \theta = 0, \quad v = 0. \quad (2.10)$$

Here P is the Prandtl number, $P = \nu/\kappa$; R is the frontal Rayleigh number, $R = (g\beta q\kappa^2)/(vc^3)$, $\delta = R_0T_b/E$, $\theta_0 = (T_b - T_0)/q$, and $\mu = \frac{2\kappa}{c^2}\sigma$.

In the next section we reduce the problem to an interface problem. In the following sections we fulfil its stability analysis.

3. Approximation of infinitely narrow reaction zone

To study the problem analytically we reduce it to a singular perturbation problem where the reaction zone is supposed to be infinitely narrow, and the reaction term is neglected outside of the reaction zone [18]. This is a conventional approach for combustion problems. We perform a formal asymptotic analysis with $\varepsilon = Z^{-1} = R_0T_b^2/(qE)$ taken as a small parameter and obtain a closed interface problem. In this section we do not assume that the product is solid.

Let $\zeta(x, y, t)$ denote the location of the reaction zone in the laboratory frame reference. The new independent variable is given by

$$z_1 = z - \zeta(x, y, t).$$

We introduce new functions $\theta_1, \alpha_1, v_1, p_1$:

$$\theta(x, y, z, t) = \theta_1(x, y, z_1, t), \quad \alpha(x, y, z, t) = \alpha_1(x, y, z_1, t),$$

$$v(x, y, z, t) = v_1(x, y, z_1, t), \quad p(x, y, z, t) = p_1(x, y, z_1, t).$$

We rewrite the Equations (2.5)–(2.8) in the form (the index 1 for the new functions is omitted):

$$\frac{\partial \theta}{\partial t} - \frac{\partial \theta}{\partial z_1} \frac{\partial \zeta}{\partial t} + v \tilde{\nabla} \theta = \tilde{\Delta} \theta + Z \exp\left(\frac{\theta}{Z^{-1} + \delta \theta}\right) \phi(\alpha), \quad (3.1)$$

$$\frac{\partial \alpha}{\partial t} - \frac{\partial \alpha}{\partial z_1} \frac{\partial \zeta}{\partial t} + v \tilde{\nabla} \alpha = Z \exp\left(\frac{\theta}{Z^{-1} + \delta \theta}\right) \phi(\alpha), \quad (3.2)$$

$$\frac{\partial v}{\partial t} - \frac{\partial v}{\partial z_1} \frac{\partial \zeta}{\partial t} + (v \tilde{\nabla}) v = -\tilde{\nabla} p + P \tilde{\Delta} v + Q (1 + \lambda \sin(\mu t)) (\theta + \theta_0) \gamma, \quad (3.3)$$

$$\frac{\partial v_x}{\partial x} - \frac{\partial v_x}{\partial z_1} \frac{\partial \zeta}{\partial x} + \frac{\partial v_y}{\partial y} - \frac{\partial v_y}{\partial z_1} \frac{\partial \zeta}{\partial y} + \frac{\partial v_z}{\partial z_1} = 0, \quad (3.4)$$

where

$$\tilde{\Delta} = \frac{\partial^2}{\partial x^2} + \frac{\partial^2}{\partial y^2} + \frac{\partial^2}{\partial z_1^2} - 2\frac{\partial^2}{\partial x \partial z_1} \frac{\partial \zeta}{\partial x} - 2\frac{\partial^2}{\partial y \partial z_1} \frac{\partial \zeta}{\partial y} +$$

$$\frac{\partial^2}{\partial z_1^2} \left(\left(\frac{\partial \zeta}{\partial x} \right)^2 + \left(\frac{\partial \zeta}{\partial y} \right)^2 \right) - \frac{\partial}{\partial z_1} \left(\frac{\partial^2 \zeta}{\partial x^2} + \frac{\partial^2 \zeta}{\partial y^2} \right),$$

$$\tilde{\nabla} = \left(\frac{\partial}{\partial x} - \frac{\partial}{\partial z_1} \frac{\partial \zeta}{\partial x}, \frac{\partial}{\partial y} - \frac{\partial}{\partial z_1} \frac{\partial \zeta}{\partial y}, \frac{\partial}{\partial z_1} \right), \quad \text{and} \quad Q = PR.$$

We use matched asymptotic expansions and seek the outer solution of the problem (3.1)–(3.4) in the form of the expansion

$$\theta = \theta^0 + \varepsilon \theta^1 + \dots, \quad \alpha = \alpha^0 + \varepsilon \alpha^1 + \dots,$$

$$v = v^0 + \varepsilon v^1 + \dots, \quad p = p^0 + \varepsilon p^1 + \dots$$

To obtain the jump conditions in the reaction zone we consider the inner problem. The stretched coordinate is $\eta = z_1/\varepsilon$, $\varepsilon = Z^{-1}$. We look for the inner solution in the form of the expansion

$$\theta = \varepsilon \tilde{\theta}^1 + \dots, \quad \alpha = \tilde{\alpha}^0 + \varepsilon \tilde{\alpha}^1 + \dots, \quad (3.5)$$

$$v = \tilde{v}^0 + \varepsilon \tilde{v}^1 + \dots, \quad p = \tilde{p}^0 + \varepsilon \tilde{p}^1 + \dots, \quad \zeta = \zeta^0 + \varepsilon \zeta^1 + \dots \quad (3.6)$$

Substituting these expansions in (3.1)–(3.4), we obtain the leading-order inner problem:

$$\left(1 + \left(\frac{\partial \zeta^0}{\partial x} \right)^2 + \left(\frac{\partial \zeta^0}{\partial y} \right)^2 \right) \frac{\partial^2 \tilde{\theta}^1}{\partial \eta^2} + \exp(\tilde{\theta}^1) \phi(\tilde{\alpha}^0) = 0, \quad (3.7)$$

$$-\frac{\partial \tilde{\alpha}^0}{\partial \eta} \frac{\partial \zeta^0}{\partial t} - \frac{\partial \tilde{\alpha}^0}{\partial \eta} \left(\tilde{v}_x^0 \frac{\partial \zeta^0}{\partial x} + \tilde{v}_y^0 \frac{\partial \zeta^0}{\partial y} - \tilde{v}_\eta^0 \right) = \exp(\tilde{\theta}^1) \phi(\tilde{\alpha}^0), \quad (3.8)$$

$$\left(1 + \left(\frac{\partial \zeta^0}{\partial x} \right)^2 + \left(\frac{\partial \zeta^0}{\partial y} \right)^2 \right) \frac{\partial^2 \tilde{v}^0}{\partial \eta^2} = 0, \quad (3.9)$$

$$-\frac{\partial \tilde{v}_x^0}{\partial \eta} \frac{\partial \zeta^0}{\partial x} - \frac{\partial \tilde{v}_y^0}{\partial \eta} \frac{\partial \zeta^0}{\partial y} + \frac{\partial \tilde{v}_\eta^0}{\partial \eta} = 0. \quad (3.10)$$

The matching conditions are:

As $\eta \rightarrow +\infty$:

$$\tilde{\theta}^1 \sim \theta^1|_{z_1=+0} + \left(\frac{\partial \theta^0}{\partial z_1} \Big|_{z_1=+0} \right) \eta, \quad \tilde{\alpha}^0 \rightarrow 0, \quad \tilde{v}^0 \rightarrow v^0|_{z_1=+0}, \quad (3.11)$$

as $\eta \rightarrow -\infty$:

$$\tilde{\theta}^1 \sim \theta^1|_{z_1=-0}, \quad \tilde{\alpha}^0 \rightarrow 1, \quad \tilde{v}^0 \rightarrow v^0|_{z_1=-0}, \quad (3.12)$$

From (3.9), we obtain

$$\frac{\partial^2 \tilde{v}^0}{\partial \eta^2} = 0.$$

Thus $\tilde{v}^0(\eta)$ is a linear function of η . From the boundedness of the velocity it follows that it is identically constant. We conclude that \tilde{v}^0 does not depend on η . From this conclusion and the matching conditions it follows that the first term in the expansion of the velocity in the outer problem, v^0 is continuous.

We denote

$$s = \tilde{v}_x^0 \frac{\partial \zeta^0}{\partial x} + \tilde{v}_y^0 \frac{\partial \zeta^0}{\partial y} - \tilde{v}_\eta^0.$$

From (3.10) it follows that the function s does not depend on η .

We derive next the jump conditions for the temperature from (3.7), (3.8) in the same way as it is usually done for combustion problems. From (3.8) it follows that $\tilde{\alpha}^0$ is a monotonic function and $0 < \tilde{\alpha}^0 < 1$. Since we consider a zero-order reaction, we have $\phi(\tilde{\alpha}^0) \equiv 1$. We conclude from (3.7) that $\tilde{\theta}^1$ is also monotonic. Thus, multiplying (3.7) by $\frac{\partial \tilde{\theta}^1}{\partial \eta}$ and integrating, we obtain:

$$\left(\frac{\partial \tilde{\theta}^1}{\partial \eta} \right)^2 \Big|_{\infty} - \left(\frac{\partial \tilde{\theta}^1}{\partial \eta} \right)^2 \Big|_{-\infty} = -2A^{-1} \exp(\theta^1) \quad (3.13)$$

where

$$A = 1 + \left(\frac{\partial \zeta^0}{\partial x} \right)^2 + \left(\frac{\partial \zeta^0}{\partial y} \right)^2.$$

Subtracting (3.7) from (3.8) and integrating, we have

$$\frac{\partial \tilde{\theta}^1}{\partial \eta} \Big|_{\infty} - \frac{\partial \tilde{\theta}^1}{\partial \eta} \Big|_{-\infty} = -A^{-1} \left(\frac{\partial \xi^0}{\partial t} + s \right). \quad (3.14)$$

The two last Equations (3.13), (3.14) give the jump conditions for the temperature across the front.

Using the matching conditions and truncating the expansion as:

$$\theta^0 \approx \theta, \quad \theta^1|_{z_1=-0} \approx Z\theta|_{z_1=0}, \quad \zeta^0 \approx \zeta, \quad v^0 \approx v,$$

we can rewrite the jump conditions in the form (see [3], [4], [40] and references therein):

$$\left(\frac{\partial \theta}{\partial z_1} \right)^2 \Big|_{+0} - \left(\frac{\partial \theta}{\partial z_1} \right)^2 \Big|_{-0} = 2Z \left(1 + \left(\frac{\partial \zeta}{\partial x} \right)^2 + \left(\frac{\partial \zeta}{\partial y} \right)^2 \right)^{-1} \exp(Z\theta|_0) \quad (3.15)$$

$$\left. \frac{\partial \theta}{\partial z_1} \right|_{+0} - \left. \frac{\partial \theta}{\partial z_1} \right|_{-0} = - \left(1 + \left(\frac{\partial \zeta}{\partial x} \right)^2 + \left(\frac{\partial \zeta}{\partial y} \right)^2 \right)^{-1} \left(\frac{\partial \zeta}{\partial t} + \left(v_x \frac{\partial \zeta}{\partial x} + \frac{\partial \zeta}{\partial y} - v_z \right) \right) \Big|_{z_1=0}. \quad (3.16)$$

We recall that the jump conditions have been derived in the general case. We consider now the case of the solid product where the velocity is zero behind the reaction zone, $v \equiv 0$ for $z_1 < \zeta$. Thus, we have the following formulation of the problem:

$z_1 > \zeta$ (liquid monomer):

$$\frac{\partial \theta}{\partial t} + v \nabla \theta = \Delta \theta, \quad (3.17)$$

$$\alpha = 0, \quad (3.18)$$

$$\frac{\partial v}{\partial t} + (v \nabla) v = -\nabla p + P \Delta v + Q (1 + \lambda \sin(\mu t)) (\theta + \theta_0) \gamma, \quad (3.19)$$

$$\operatorname{div} v = 0, \quad (3.20)$$

$z_1 < \zeta$ (solid polymer):

$$\frac{\partial \theta}{\partial t} = \Delta \theta, \quad (3.21)$$

$$\alpha = 1, \quad v = 0, \quad (3.22)$$

$z_1 = \zeta$ (interface):

$$\theta|_{\zeta-0} = \theta|_{\zeta+0}, \quad (3.23)$$

$$\left. \frac{\partial \theta}{\partial z_1} \right|_{\zeta-0} - \left. \frac{\partial \theta}{\partial z_1} \right|_{\zeta+0} = \left(1 + (\zeta'_x)^2 + (\zeta'_y)^2 \right)^{-1} \frac{\partial \zeta}{\partial t}, \quad (3.24)$$

$$\left(\frac{\partial \theta}{\partial z_1} \right)^2 \Big|_{\zeta-0} - \left(\frac{\partial \theta}{\partial z_1} \right)^2 \Big|_{\zeta+0} = -2Z \left(1 + (\zeta'_x)^2 + (\zeta'_y)^2 \right)^{-1} \exp(Z \theta|_{\zeta}) \quad (3.25)$$

$$v_x = v_y = v_z = 0. \quad (3.26)$$

The conditions at infinity are

$$z_1 = -\infty : \theta = 0, \quad v = 0; \quad z_1 = +\infty : \theta = -1, \quad v = 0. \quad (3.27)$$

The problem is coupled in the sense that it describes the thermal instability of the reaction front and the convective instability at the same time. There are different limiting cases here. For example, if the coefficient of thermal expansion, β , is zero (*i.e.*, $R = 0$) then we have,

obviously, a pure problem of the reaction in a condensed medium, since $v \equiv 0$. Another limiting case is when we remove the thermal instability: if the Zeldovich number is less than a critical value, then there is no thermal instability, and it can be treated as a purely convective instability.

4. Linear stability analysis

In this section we perform the linear analysis of the steady-state solution for the interface problem (3.17)–(3.27).

This problem has a travelling wave solution:

$$\theta(x, y, z_1, t) = \theta_s(z_1 - ut), \quad \alpha(x, y, z_1, t) = \alpha_s(z_1 - ut), \quad v = 0,$$

where

$$\theta_s(z_2, t) = \begin{cases} 0 & \text{if } z_2 < 0 \\ \exp(-uz_2) - 1 & \text{if } z_2 > 0 \end{cases}, \quad (4.1)$$

and

$$\alpha_s(z_2, t) = \begin{cases} 1 & \text{if } z_2 < 0 \\ 0 & \text{if } z_2 > 0 \end{cases}, \quad (4.2)$$

$$z_2 = z_1 - ut, \quad u = c.$$

This is a stationary solution of the problem given by the Equations (3.18), (3.20), (3.22)–(3.27) and

$$\frac{\partial \theta}{\partial t} + v \nabla \theta = \Delta \theta + u \frac{\partial \theta}{\partial z_2}, \quad (4.3)$$

$$\frac{\partial v}{\partial t} + (v \nabla) v = -\nabla p + P \Delta v + u \frac{\partial v}{\partial z_2} + Q(1 + \lambda \sin(\mu t))(\theta + \theta_0) \gamma, \quad (4.4)$$

for $z_2 > \xi$,

$$\frac{\partial \theta}{\partial t} = \Delta \theta + u \frac{\partial \theta}{\partial z_2}, \quad (4.5)$$

for $z_2 < \xi$. Here $\xi = \zeta - ut$.

We study the stability of this solution. We look for a solution of the problem in the form of the perturbed stationary solution:

$$\theta = \theta_s + \tilde{\theta}, \quad v = v_s + \tilde{v}, \quad p = p_s + \tilde{p}. \quad (4.6)$$

We substitute (4.6) in (3.20), (4.3)–(4.5) and obtain for the first-order terms:

$z_2 > \xi$:

$$\frac{\partial \tilde{\theta}}{\partial t} = \Delta \tilde{\theta} + u \frac{\partial \tilde{\theta}}{\partial z_2} - \tilde{v}_z \theta_s', \quad (4.7)$$

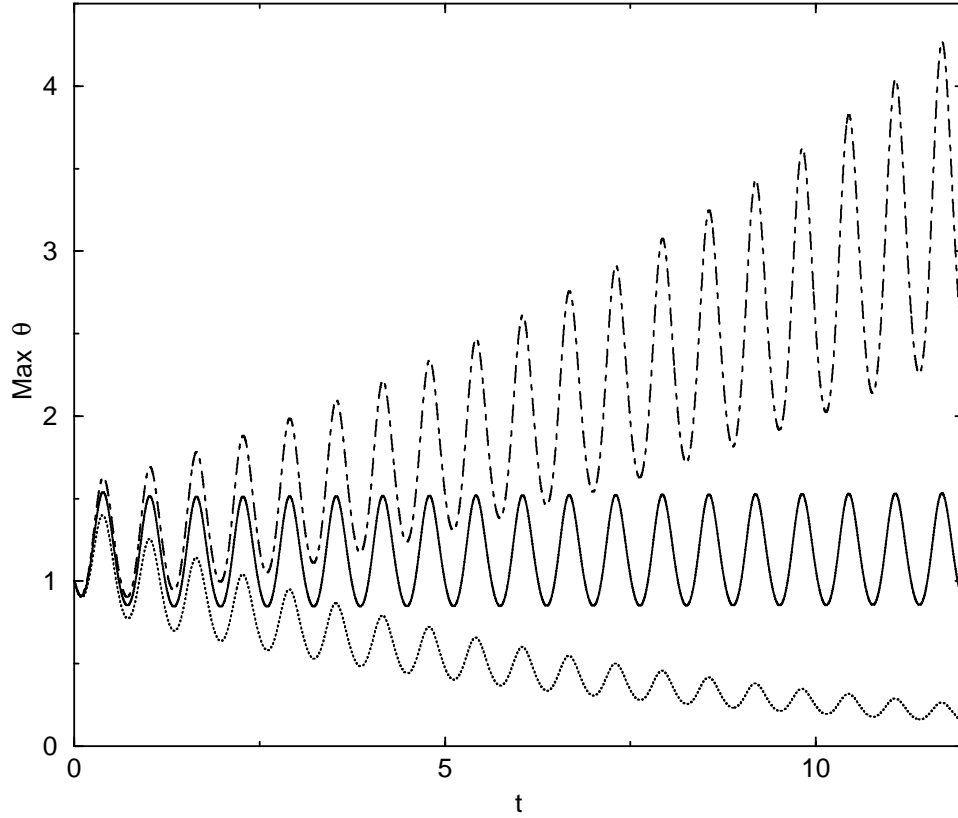


Figure 2. Temperature maximum as a function of time for $\lambda = 2$, $k = 1.5$, $P = 10$ and $\mu = 10$ (... $R = 85$, — $R = 91.3$ and --- $R = 95$).

$$\frac{\partial \tilde{v}}{\partial t} = -\nabla \tilde{p} + P \Delta \tilde{v} + u \frac{\partial \tilde{v}}{\partial z_2} + Q(1 + \lambda \sin(\mu t)) \tilde{\theta} \gamma, \quad (4.8)$$

$$\operatorname{div} \tilde{v} = 0,$$

$z_2 < \xi$:

$$\frac{\partial \tilde{\theta}}{\partial t} = \Delta \tilde{\theta} + u \frac{\partial \tilde{\theta}}{\partial z_2}. \quad (4.9)$$

We denote $\tilde{\theta} = \hat{\theta}_1$ for $z_2 < \xi$ and $\tilde{\theta} = \hat{\theta}_2$ for $z_2 > \xi$.

We now linearize the jump conditions. Taking into account that

$$\theta|_{\xi \pm 0} = \theta_s(0) + \xi \theta'_s(\pm 0) + \tilde{\theta}(\pm 0),$$

$$\left. \frac{\partial \theta}{\partial z_2} \right|_{\xi \pm 0} = \theta'_s(\pm 0) + \xi \theta''_s(\pm 0) + \left. \frac{\partial \tilde{\theta}}{\partial z_2} \right|_{\xi \pm 0},$$

we obtain up to the higher-order terms

$$[\hat{\theta}] = u\xi, \quad (4.10)$$

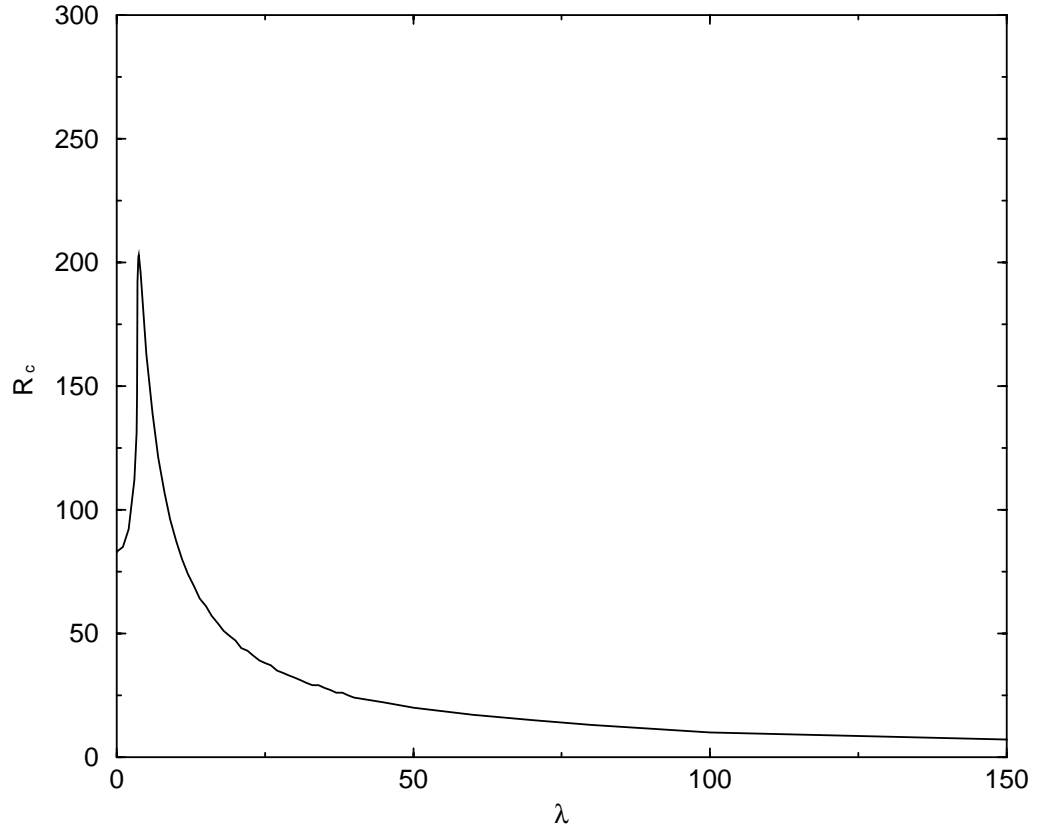


Figure 3. Convective instability boundary: critical Rayleigh number as a function of the amplitude of vibrations for $P = 10$, $k = 1.5$ and $\mu = 10$.

$$[\hat{\theta}'] = -u^2\xi - \frac{d\xi}{dt}, \quad (4.11)$$

$$-u \left(u^2\xi + \hat{\theta}'_2(0) \right) = Z\hat{\theta}'_1(0). \quad (4.12)$$

Here

$$[\hat{\theta}] = \hat{\theta}_2(0) - \hat{\theta}_1(0), \quad [\hat{\theta}'] = \hat{\theta}'_2(0) - \hat{\theta}'_1(0), \quad \hat{\theta}'_i(0) = \left. \frac{\partial \hat{\theta}_i}{\partial z_2} \right|_{z_2=0}.$$

From (3.26) we have

$$\frac{\partial v_x}{\partial x} + \frac{\partial v_x}{\partial z} \frac{\partial \xi}{\partial x} = 0, \quad \frac{\partial v_y}{\partial y} + \frac{\partial v_y}{\partial z} \frac{\partial \xi}{\partial y} = 0.$$

Thus, using (3.20) we can rewrite the boundary conditions for the first-order terms in the form

$$\hat{v}_z = 0, \quad \frac{\partial \hat{v}_z}{\partial z} = 0. \quad (4.13)$$

We consider the perturbations in the form:

$$\hat{\theta}_i = \theta_i(z_2, t) \exp(i(k_1x + k_2y)), \quad i = 1, 2, \quad (4.14)$$

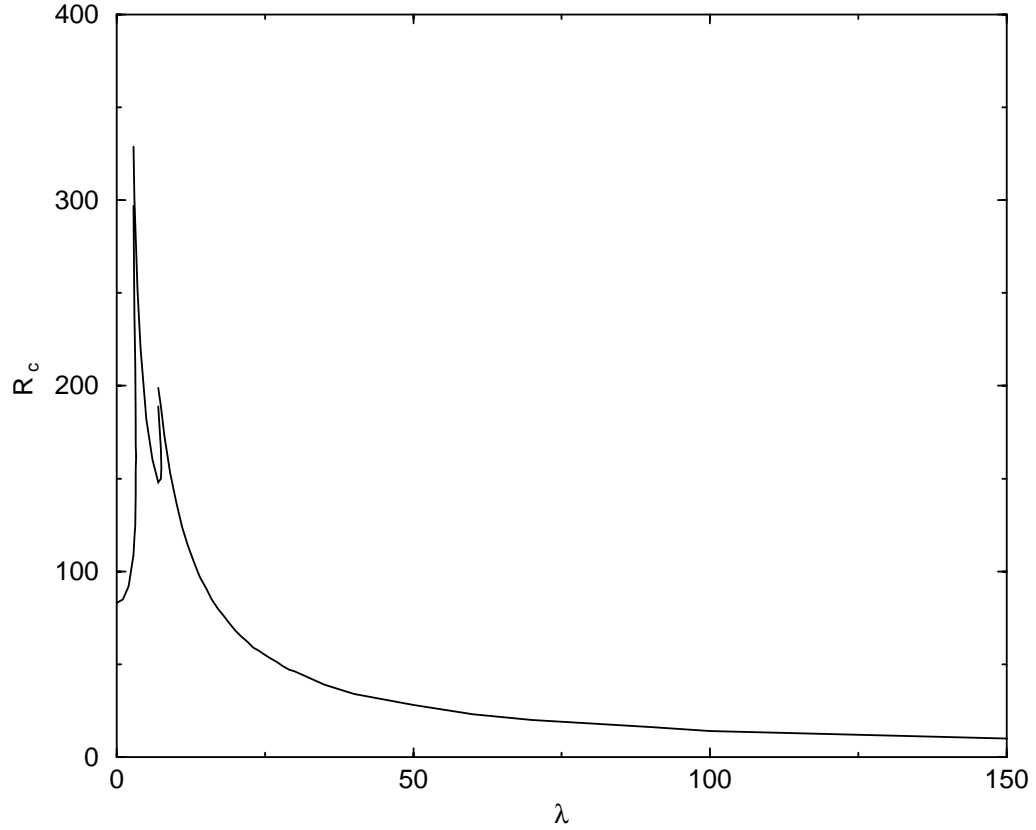


Figure 4. Convective instability boundary: critical Rayleigh number as a function of the amplitude of vibrations for $P = 10$, $k = 1.5$ and $\mu = 15$.

$$\tilde{v} = v_2(z_2, t) \exp(i(k_1x + k_2y)), \quad z_2 > \xi, \quad (4.15)$$

$$\xi = \varepsilon_1(t) \exp(i(k_1x + k_2y)). \quad (4.16)$$

Here ε_1 is the amplitude, k_1 and k_2 the wave numbers. We exclude the pressure p and the components v_x and v_y of the velocity from the system (4.3)–(4.5) applying two times the operator *curl* to the Navier-Stokes equations. Thus we can rewrite (4.3), (4.4) in the form

$$\frac{\partial \tilde{\theta}}{\partial t} - u \frac{\partial \tilde{\theta}}{\partial z_2} + \tilde{v}_z \theta'_s = \Delta \tilde{\theta}, \quad (4.17)$$

$$\frac{\partial}{\partial t} \Delta \tilde{v}_z - u \frac{\partial}{\partial z_2} \Delta \tilde{v}_z = P \Delta \Delta \tilde{v}_z + Q \left(\frac{\partial}{\partial x^2} + \frac{\partial}{\partial y^2} \right) \tilde{\theta} (1 + \lambda \sin(\mu t)). \quad (4.18)$$

Substituting (4.14), (4.15) in (4.17), (4.18) we obtain the system

$$\frac{\partial \theta}{\partial t} = \theta'' + u\theta' - k^2\theta + u \exp(-uz_2)v, \quad (4.19)$$

$$\frac{\partial}{\partial t} (v'' - k^2v) = P (v'''' - 2k^2v'' + k^4v) + u (v''' - k^2v') - PRk^2 (1 + \lambda \sin(\mu t)) \theta \quad (4.20)$$

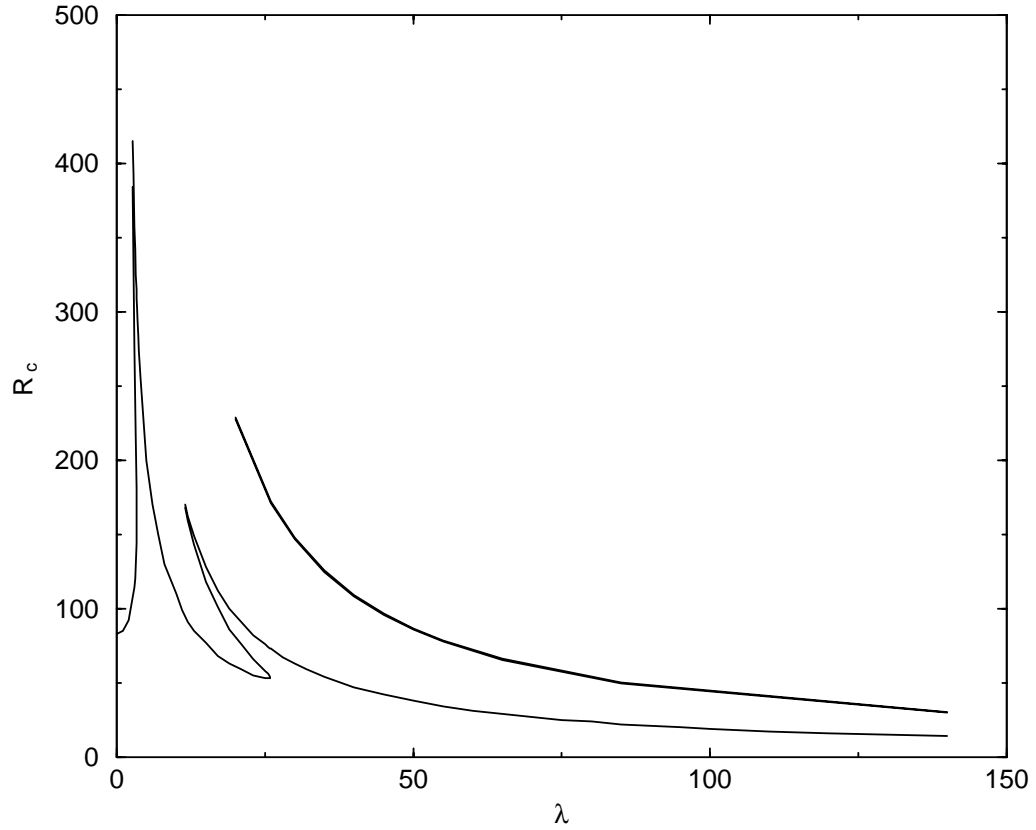


Figure 5. Convective instability boundary: critical Rayleigh number as a function of the amplitude of vibrations for $P = 10$, $k = 1.5$ and $\mu = 20$.

with the boundary conditions:

$$v(0, t) = v'(0, t) = 0, \quad (4.21)$$

$$\theta'(0, t) = -u\theta(0, t). \quad (4.22)$$

Here $k = \sqrt{k_1^2 + k_2^2}$.

The boundary condition for the velocity follows directly from (4.13). To obtain (4.22), we note first of all that the temperature perturbation behind the front, where the medium is unmovable, satisfies the equation

$$\frac{\partial \theta_1}{\partial t} = \theta_1'' + u\theta_1' - k^2\theta_1.$$

We look for the solution in the form

$$\theta_1(z_2, t) = \theta_1(z_2)f(t)$$

and obtain

$$\theta_1(z_2, t) = c_1 e^{\omega t} e^{\mu_1 z_2}, \quad \mu_1 = -\frac{u}{2} - \sqrt{\frac{u^2}{4} + k^2 + \omega}.$$

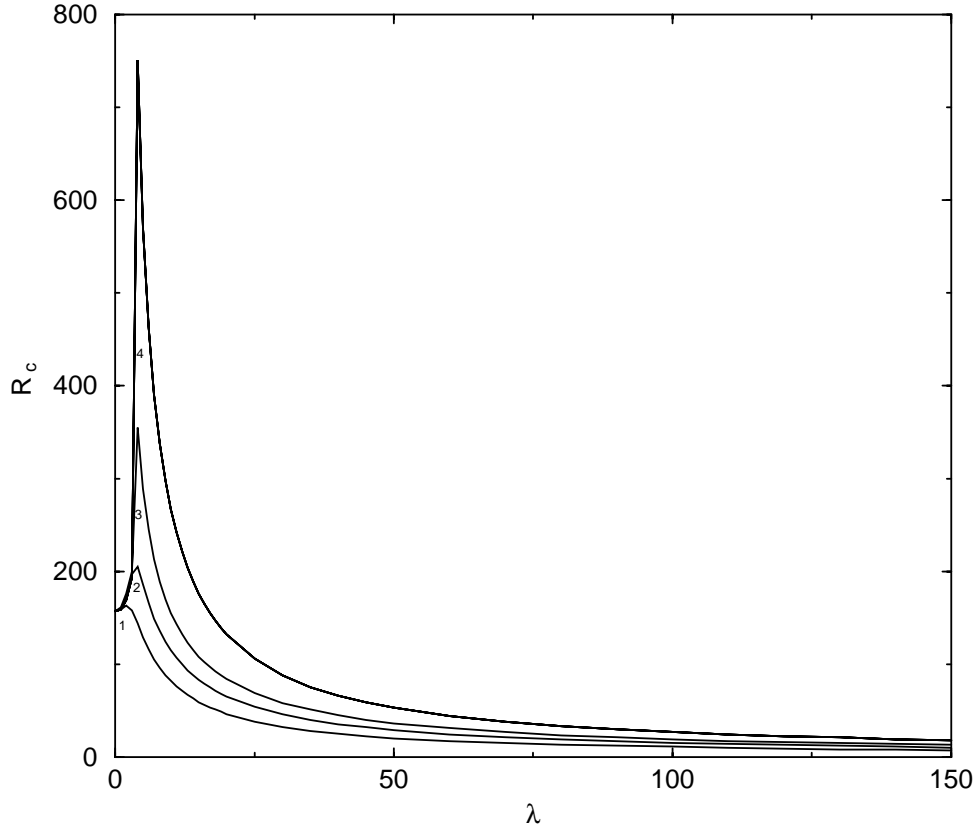


Figure 6. Convective instability boundary: critical Rayleigh number as a function of the amplitude of vibrations for $P = 10$, $k = 2$, $1.\mu = 5$, $2.\mu = 10$, $3.\mu = 15$ and $4.\mu = 60$.

We recall that there are two basic types of instability. In the first case a pair of complex conjugate eigenvalues cross the imaginary axis resulting in a Hopf bifurcation. In the second case, which we consider here, an eigenvalue crosses the imaginary axis through zero. Hence in this case $\omega = 0$ at the stability boundary.

We obtain from (4.11), (4.12) for $\omega = 0$:

$$-u \left(\mu_1 c_1 - \frac{d\xi}{dt} \right) = Z c_1.$$

Since two terms in this equation do not depend on t , we conclude that $\xi(t) = \text{const.}$

Thus from the same equation

$$c_1 (Z + u\mu_1) = 0.$$

Since Z is a large parameter and $u \approx 1$, thus generally speaking $Z \neq -u\mu_1$ and, consequently, $c_1 = 0$. From (4.10), (4.11) we obtain (4.22).

This analysis allows us to conclude that there is no temperature perturbation behind the front, and that the reaction zone remains unmovable. This conclusion was first drawn in [15] for the case without vibrations. We see that it remains valid for the case with vibrations though *a priori* it is not clear whether it is so.

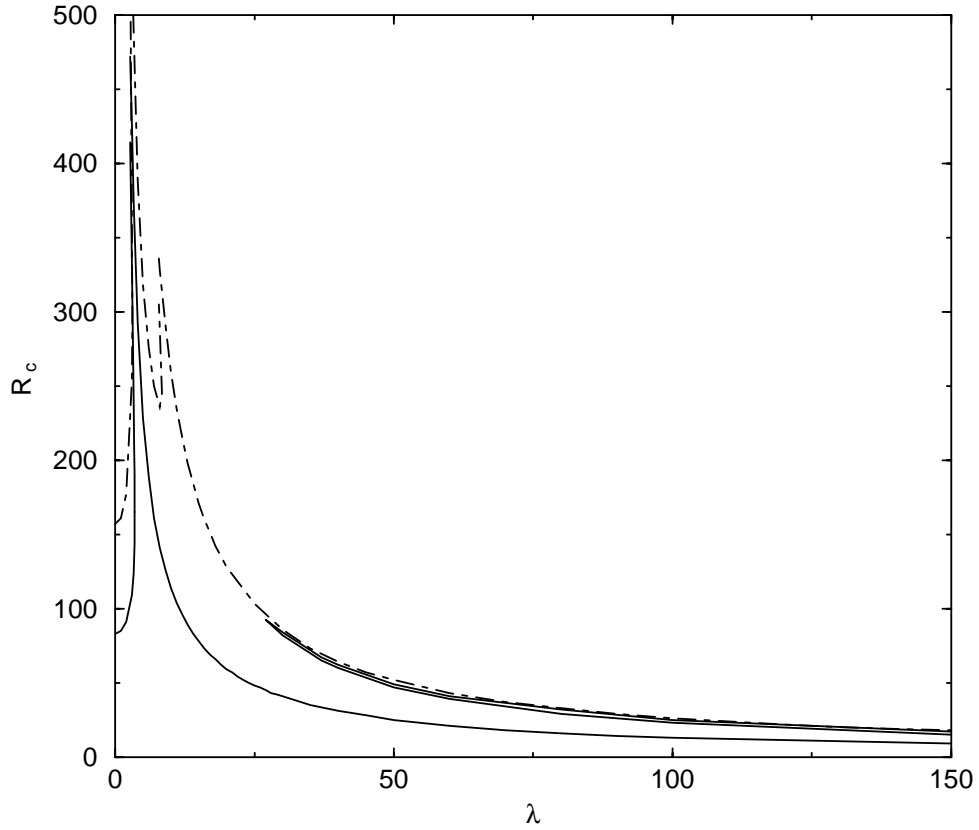


Figure 7. Convective instability boundary: critical Rayleigh number as a function of the amplitude of vibrations for $P = 10$, $\mu = 25$, (— $k = 1.5$, --- $k = 2$).

5. Numerical results

To find the convective instability boundary, we solve numerically the problem (4.19) – (4.22). We describe briefly the numerical method. We reduce the problem (4.19) – (4.22) to the problem

$$\frac{\partial \theta}{\partial t} = \theta'' + u\theta' - k^2\theta + u \exp(-uz)v, \quad (5.1)$$

$$\frac{\partial \omega}{\partial t} = P\omega'' + u\omega' - Pk^2\omega + PRk^2(1 + \varepsilon \sin(\mu t))\theta$$

$$0 = v'' - k^2v + \omega,$$

in the interval $0 \leq x \leq L$, with the boundary conditions

$$x = 0 : \quad \theta' = -u\theta, \quad v = v' = 0,$$

$$x = L : \quad \theta = v = \omega = 0.$$

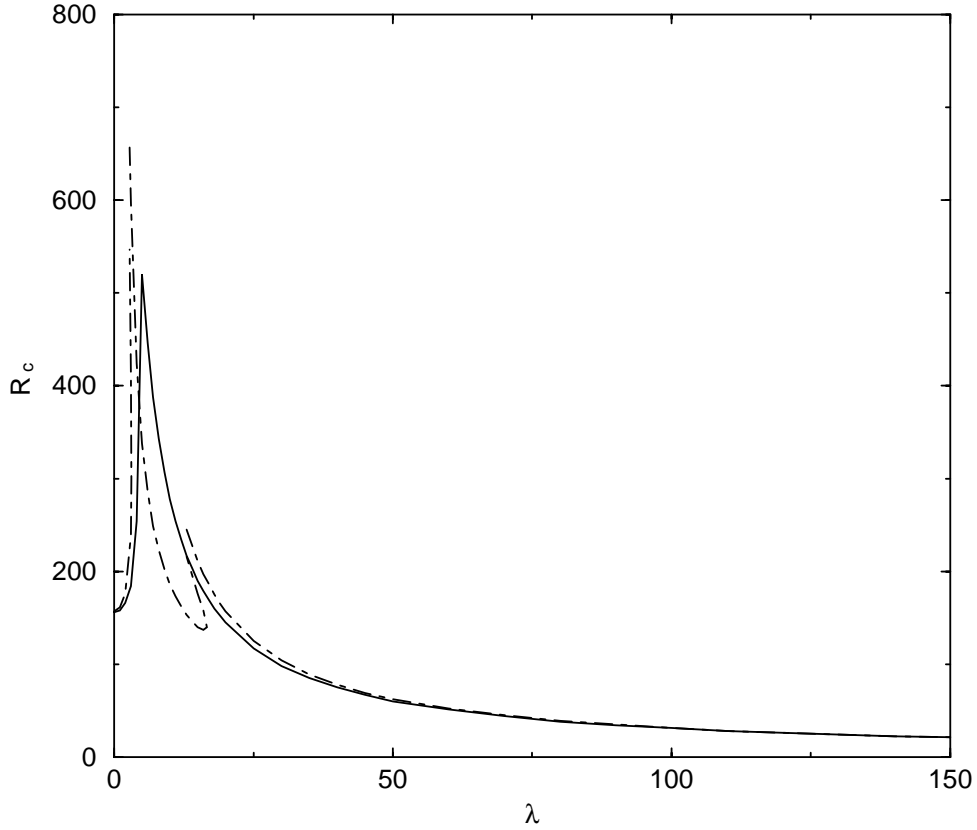


Figure 8. Convective instability boundary: critical Rayleigh number as a function of the amplitude of vibrations for $k = 2$, $\mu = 30$ (--- $P = 10$, — $P = 20$).

We note that there are two boundary conditions for the velocity at $x = 0$ and there is no boundary condition for the vorticity ω . To avoid this difficulty we use a conventional approach introducing an artificial second-order boundary condition for the vorticity (see [41]):

$$2\omega_1 + \omega_2 = -\frac{6v_2}{h^2}. \quad (5.2)$$

Here the index 1 corresponds to the first discretization point and 2 to the second points; h is the space step. We use a finite-difference approximation with implicit scheme except for the boundary condition (5.2) where the velocity v is taken from the previous time step. The numerical accuracy is controlled by decreasing the time and space steps. We verify that the results do not depend on the length L of the interval.

For fixed Z and P we vary R . If the Rayleigh number R is less than a critical value R_{cr} , then solution is decreasing in time (Figure 2). If $R > R_{\text{cr}}$, it increases, and for $R = R_{\text{cr}}$ it is periodic in time.

Figures 3 – 5 show the critical value of the Rayleigh number as a function of the amplitude of vibrations for different frequencies. If $\lambda = 0$, we obtain the same value $R_{\text{cr}} \approx 70$ as without vibrations [15, 16]. For small positive λ vibrations stabilize the solution: R_{cr} is an increasing function. For larger λ there appears a decreasing branch of the stability boundary (Figure 3).

It corresponds to a parametric instability where vibrations with high amplitude destabilize the front.

We note that these two branches have the same asymptotic behavior for large R but they do not coincide. It can be seen also in Figure 4 where there are already three branches of the stability boundary. The intermediate branch descends as μ increases (Figure 5). For this value of frequency we have found also a very narrow stability region located between two almost coinciding curves. It is connected with the main stability region for large values of λ . In terms of eigenvalues, the interpretation of this diagram is as follows. If we fix λ and increase R from zero, then the stationary solution loses its stability when R crosses the stability boundary for the first time. For this value of R , the eigenvalue with the maximal real part crosses 0. For larger values of R it crosses 0 again, but this time from the right-half plane to the left-half plane. The stationary solution becomes stable. Finally it crosses 0 for the third time, which makes the solution unstable again.

Figure 6 shows the stability boundary for another value of k . Increase of frequency of vibrations makes the front more stable.

Dependence of the critical Rayleigh number on the wave number is shown in Figure 7. We can see that main parametric instability region remains practically the same. For $k = 2$ and large R there is another parametric instability region. For $k = 1.5$ this region increases and leaves a very narrow strip near the main instability domain where the front is stable.

We discuss finally dependence of the stability conditions on the Prandtl number. We note that two terms in (4.20) depend on it. An increase P in the first term corresponds to increasing the viscosity which has a stabilizing effect. Increasing of P in the last term corresponds to an increase of the external force, which destabilizes the front.

In the case without vibrations the sum of these two phenomena make the front less stable. However, for $P > 2$ the stability boundary is practically independent of it [16].

In the case with vibrations the convective instability region (small amplitudes) increases with P and the front is less stable (Figure 8). The main parametric instability region also increases. However for $P = 10$ there is another parametric instability region, which appears between the convective and the main parametric instability regions.

6. Conclusion

In this work we have studied the influence of vibrations on the convective instability of a polymerization front with a liquid monomer and a solid polymer. For the front propagating upwards, the exothermic chemical reaction heats the liquid reactant from below. If the frontal Rayleigh number exceeds a critical value, the plane front loses its stability, and a propagating convective front bifurcates. This phenomenon is studied experimentally in [13, 14].

Vibrations can change the onset of convection. To study their influence we consider the model consisting of reaction-diffusion equations coupled with the Navier-Stokes equations under the Boussinesq approximation. Vibrations are taken into account through the buoyancy term, where the gravity acceleration depends periodically on time.

For a linear stability analysis we use the infinitely narrow reaction zone method developed by Zeldovich and Frank-Kamenetski [18] and used for various combustion problems (see [1–7]). In this method it is assumed that the reaction occurs at a surface separating original reactants and the product of the reaction. The nonlinear reaction term is neglected outside the reaction zone, and the problem is completed by nonlinear jump conditions. The moving-

boundary problem has a travelling-wave solution that can be found explicitly. Linearizing the problem about it, we obtain an eigenvalue problem with time-dependent coefficients. We solve it numerically and find the stability boundary.

The stability conditions can be formulated in terms of the frontal Rayleigh number. If it exceeds a critical value R_c the convection appears. The critical Rayleigh number depends on the amplitude λ and on the frequency μ of vibrations. For small amplitudes R_c increases with λ , *i.e.*, vibrations stabilize the front. The structure of the stability region is rather complex: there exists a narrow stability region even for a very high values of the Rayleigh number.

For larger amplitudes there appears another branch of the stability boundary. It corresponds to the so-called parametric instability, and it decreases with λ . The parametric instability is also very sensitive to parameters. The corresponding stability boundary can contain several disjoint parts.

Increasing of the frequency of vibrations stabilizes the front with respect to the convective instability (first branch of the stability boundary). The dependence of the parametric instability on the frequency is complex.

We note finally that the case where the polymer is liquid is essentially different from the case of a solid polymer considered in this work. The most essential difference is that the convective instability exists for descending fronts and not only for ascending ones as in the case of a solid product of reaction [15]. The influence of vibrations in this case has not yet been studied and it can be also different.

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