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Reaction fronts in porous media, influence of Lewis number, linear stability analysis

Research Article

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Abstract: This work is devoted to the investigation of propagating polymerization fronts in porous media. We consider a simplified mathematical model which consists of coupling two convection-diffusion-reaction equations for the temperature and depth of conversion to the Darcy equation for the pressure. A formal asymptotic analysis of the problem is carried out taking into account the Zeldovich - Frank-Kamanetskii approximation. We fulfill the linear stability analysis of the stationary propagating front and show that the Lewis number influences conditions of the convective instability.

MSC: 35K57 • 76S05

Keywords: Frontal polymerization • Porous media • Lewis number • Zeldovich - Frank-Kamanetskii approximation • Linear stability analysis

1. Introduction

Frontal polymerization is a mode of converting monomer (reactant) into polymer (product) via a localized reaction zone that propagates through the coupling of thermal diffusion and Arrhenius reaction kinetics.

In many cases propagation of polymerization fronts is accompanied by various physical phenomena and instabilities. In [1, 2], it is shown that the front is influenced by the thermo-diffusional instability. It's induced by the disparities between mass and thermal diffusivities of a combustible mixture. These works are completed by [3, 4] where the process is affected by the interaction of reacting gases or liquids with a porous matrix resulting in fingering of chemical reaction fronts, and also by some specific phenomena such as filtration combustion [5–7]. In case the density of the medium depends on the temperature, its expansion in the reaction zone may affect the hydrodynamic instability. It's called Darrieus-Landau instability which is investigated in [8, 9]. In other studies like [10–13], the focus is about the convective instability which can appear due to natural convection, once the frontal Rayleigh number is sufficiently large or under the effect of vibrations. This is analysed in the pure form by taking into account the Boussinesq approximation. Most recently, some studies have identified new factors that enhance understanding of front instability. For example, it is demonstrated in [14] that variable gravity field could have a significant impact on the thermal instability. Then, in presence of thermal radiation and viscous dissipation, it is shown in [15] that the effect of various parameters like the magnetic field parameter, radiation parameter and Prandtl number, on the mixed convection heat and mass transfer of radiating fluid is important in context of processes involving high temperature.

The present work is primarily motivated by the above investigations. We consider the problem of an ascending frontal polymerization in a porous media, in the case when the reactant and the product are liquids. The main purpose

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is to study the influence of Lewis number $L_e = \kappa/d$ on the convective instability of the reaction front, where κ is the thermal diffusivity and d is the mass diffusivity.

Therefore we present in section 2, the governing equations which consists of the heat equation, the equation for the depth of conversion and the equations of motion under the Darcy law. In section 3, we perform a formal asymptotic analysis under the Zeldovich and Frank-Kamenetskii approach [2] : Most polymerization processes are exothermic. They are characterized by the fact that the basic chemical transformation takes place over a narrow temperature interval that is close to the maximum temperature. This has enabled Zeldovich and Frank-Kamenetskii to propose the infinitely narrow reaction zone method in which it is assumed that the reaction zone is concentrated at a point, and outside of this reaction zone, the non-linear source is set equal to zero. This makes it possible to replace the non-linear differential equations by linear equations and algebraic matching conditions across the reaction zone. In section 4, we present the formulation of a closed interface problem. Finally, in section 5, we carry out the linear stability analysis in order to obtain the dispersion relation. So, the stability conditions are deduced in the case of the cellular instability.

2. Governing Equations

An illustration of frontal polymerization in liquids is shown in Fig. 1. In this case the monomer and the polymer are separated by a narrow reaction zone (thermal front) that propagates if the reaction is exothermic and highly activated. We consider an upward propagation through a porous media in a cylindrical tube, which is in the direction opposite

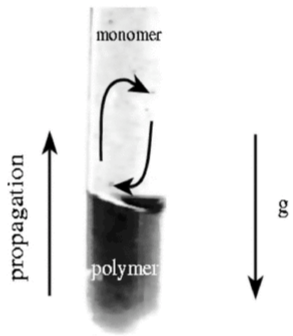

Fig. 1. Ascending propagation of polymerization front with convection

to the gravity. The tube is long, and its radius is large enough that we can neglect the walls.

The fluid is Newtonian and all the thermophysical properties are supposed to be constant, except for the density in the buoyancy term that can be adequately modelled by the Boussinesq approximation, and that compression effects and viscous dissipation are neglected. With these assumptions, the problem is described in a two-dimensional space by the following equations:

$$\frac{\partial T}{\partial t} + \mathbf{v} \bullet \nabla T = \kappa \Delta T + qK(T)\phi(\alpha), \tag{1}$$

$$\frac{\partial \alpha}{\partial t} + \mathbf{v} \bullet \nabla \alpha = d\Delta \alpha + K(T)\phi(\alpha), \tag{2}$$

$$\mathbf{v} + \frac{K}{\mu}\nabla p = \frac{g\beta K_p}{\mu}\rho(T - T_0)\gamma, \tag{3}$$

$$\nabla \bullet \mathbf{v} = 0, \tag{4}$$

with the boundary conditions

$$y \to (+\infty, -\infty) \;\; (T, \alpha, \mathbf{v}) = ((T_i, T_b), (0,1), (0,0)). \tag{5}$$

Here, T is the temperature of the combustible mixture, α the depth of conversion, p the pressure, $\mathbf{v} = (v_x, v_y)$ the fluid velocity, κ the coefficient of thermal diffusivity, q the adiabatic heat release, d the diffusion coefficient, K_p the permeability of the porous media, μ the viscosity, g the gravity acceleration, β the coefficient of thermal expansion,

ρ is the density and γ is the unit vector in the vertical direction (upward). ∇ and Δ denote the standard differential operators defined by $\nabla = \left(\frac{\partial}{\partial x}, \frac{\partial}{\partial y}\right)$ and $\Delta = \frac{\partial^2}{\partial x^2} + \frac{\partial^2}{\partial y^2}$, where (x, y) are the space coordinates. T_0 indicates the mean value of the temperature, T_i is an initial temperature while T_b is the temperature of the reacted mixture given by $T_b = T_i + q$.

The function $K(T)\phi(\alpha)$ describes the reaction rate where the temperature dependence is given by the Arrhenius law $K(T) = k_0 \exp\left(-\frac{E}{R_0 T}\right)$, k_0 is the pre-exponential factor, R_0 the universal gas constant and E is the activation energy, supposed to be large in this problem. $\phi(\alpha)$ is the kinetic function for which we consider the zero order reaction for the asymptotic analysis,

$$\phi(\alpha) = \begin{cases} 1 \text{ if } \alpha < 1 \\ 0 \text{ if } \alpha = 1 \end{cases} . \tag{6}$$

The heat release by the exothermic chemical reaction results a non-homogeneous temperature distribution and can lead to a convective instability.

If the medium is at rest, this means that $\mathbf{v} = 0$, then there exists a stationary propagating front with a velocity and a temperature distribution that can be found asymptotically for large values of the Zeldovich number
$Z = qE/R_0 T_b^2$.
In the case of the zero order reaction we have

$$c^2 = \frac{2k_0\kappa}{q}\frac{R_0 T_b^2}{E}\exp\left(\frac{-E}{R_0 T_b}\right), \tag{7}$$

$$(T, \alpha) = \begin{cases} (T_b, 1) & \text{if } \bar{y} < 0 \\ (T_i + (T_b - T_i)\exp\left(\frac{-\bar{y}c}{\kappa}\right), 0) & \text{if } \bar{y} > 0 \end{cases} . \tag{8}$$

We now introduce the following spatial variables, in order to obtain the dimensionless model:

$$x' = xc_1/\kappa,\ y' = yc_1/\kappa,\ t' = tc_1^2/\kappa,\ \mathbf{v}' = \mathbf{v}/c_1 \quad \text{and} \quad p' = p\frac{K_p}{\kappa\mu} \text{ with } c_1 = c/\sqrt{2}.$$

Denoting $\theta = (T - T_b)/q$ and keeping for convenience the same notation for the other variables, we may re-write system (1)-(4) as follows:

$$\frac{\partial\theta}{\partial t} + \mathbf{v}\bullet\nabla\theta = \Delta\theta + W_Z(\theta)\phi(\alpha), \tag{9}$$

$$\frac{\partial\alpha}{\partial t} + \mathbf{v}\bullet\nabla\alpha = L_e^{-1}\Delta\alpha + W_Z(\theta)\phi(\alpha), \tag{10}$$

$$\mathbf{v} + \nabla p = R_p(\theta + \theta_0)\gamma, \tag{11}$$

$$\nabla\bullet\mathbf{v} = 0, \tag{12}$$

with the boundary conditions

$$y \to (+\infty, -\infty)\ (\theta, \alpha, \mathbf{v}) = ((-1, 0), (0, 1), (0, 0)). \tag{13}$$

Here $W_Z(\theta) = Z\exp\left(\frac{\theta}{Z^{-1} + \delta\theta}\right)$ and the function ϕ is defined in (6).

$L_e = \kappa/d$ is the Lewis number, $R_p = K_p c_1^2 P^2 R\rho/\mu^2$ where R is the Rayleigh number and P is the Prandtl number defined by $R = g\beta q\kappa^2/\mu c_1^3$ and $P = \mu/\kappa$, the parameters δ and θ_0 are defined by $\delta = R_0 T_b/E$ and $\theta_0 = (T_b - T_0)/q$.
If we substrate (10) from (9), we obtain the following equation:

$$\frac{\partial(\theta - \alpha)}{\partial t} + \mathbf{v}\bullet\nabla(\theta - \alpha) = \Delta(\theta - L_e^{-1}\alpha), \tag{14}$$

where $\theta - \alpha = S$ is known as the enthalpy.

3. Approximation of infinitely narrow reaction zone

In this section, we present the asymptotic analysis of the problem based on the narrow reaction zone method. Indeed, for large Zeldovich number Z, if the width of the reaction zone is small and finite, the asymptotic solution can be sought in the form of an expansion in a small parameter $\epsilon = 1/Z$ connected with the width of the reaction zone. Moreover, we assume that the Lewis number can be written in the form of an expansion in ϵ, so that $L_e = 1 + \frac{1}{Z} l_e$ and $L_e^{-1} = 1 - \frac{1}{Z} l_e$ as considered by Pelaez J. and Linan A. in [16].

3.1. Outer solution

We look for the outer solution of the problem in the form of expansion

$$\begin{aligned} &\theta = \theta^0 + \epsilon\theta^1 + \ldots, \quad \alpha = \alpha^0 + \epsilon\alpha^1 + \ldots, \\ &\mathbf{v} = \mathbf{v}^0 + \epsilon\mathbf{v}^1 + \ldots, \quad p = p^0 + \epsilon p^1 + \ldots. \end{aligned} \tag{15}$$

Here $(\theta^0, \alpha^0, \mathbf{v}^0)$ is the dimensionless form of the basic solution given in (7)-(8).
Order ϵ^0

$$\frac{\partial \theta^0}{\partial t} + \mathbf{v}^0 \bullet \nabla\theta^0 = \Delta\theta^0, \tag{16}$$

$$\frac{\partial \alpha^0}{\partial t} + \mathbf{v}^0 \bullet \nabla\alpha^0 = \Delta\alpha^0, \tag{17}$$

$$\mathbf{v}^0 + \nabla p^0 = R_p(\theta^0 + \theta_0)\gamma, \tag{18}$$

$$\nabla \bullet \mathbf{v}^0 = 0, \tag{19}$$

$$\frac{\partial(\theta^0 - \alpha^0)}{\partial t} + \mathbf{v}^0 \bullet \nabla(\theta^0 - \alpha^0) = \Delta(\theta^0 - \alpha^0), \tag{20}$$

with the boundary conditions

$$y \to +\infty,\ \theta^0 = -1,\ \alpha^0 = 0 \text{ and } \mathbf{v}^0 = 0,\ y \to -\infty,\ \theta^0 = 0,\ \alpha^0 = 1 \text{ and } \mathbf{v}^0 = 0. \tag{21}$$

If it's supposed that $(\theta - \alpha)_{t=0} = -1 + O(\epsilon)$, taking account to this assumption with the boundary conditions (21), the equation (20) admits as unique solution:

$$\theta^0 - \alpha^0 = -1. \tag{22}$$

Order ϵ^1

$$\frac{\partial \theta^1}{\partial t} + \mathbf{v}^1 \bullet \nabla\theta^0 + \mathbf{v}^0 \bullet \nabla\theta^1 = \Delta\theta^1, \tag{23}$$

$$\frac{\partial \alpha^1}{\partial t} + \mathbf{v}^1 \bullet \nabla\alpha^0 + \mathbf{v}^0 \bullet \nabla\alpha^1 = \Delta\alpha^1 - l_e\Delta\alpha^0, \tag{24}$$

$$\mathbf{v}^1 + \nabla p^1 = R_p\theta^1\gamma, \tag{25}$$

$$\nabla \bullet \mathbf{v}^1 = 0. \tag{26}$$

If we substrate (24) from (23) and take into account (22), we may obtain the following equation:

$$\frac{\partial S}{\partial t} + \mathbf{v}^0 \bullet \nabla S = \Delta S + l_e\Delta\alpha^0, \tag{27}$$

where $S = \theta^1 - \alpha^1$.

3.2. Inner solution

We now consider $\zeta(t,x)$, as a small displacement of the location of the reaction zone, and we define the new independent variable $y_1 = y-\zeta(t,x)$. We introduce new unknown functions θ_1, α_1, $\mathbf{v}_1$, p_1:

$$\begin{aligned}&\theta(t,x,y)=\theta_1(t,x,y_1),\ \ \alpha(t,x,y)=\alpha_1(t,x,y_1),\\ &\mathbf{v}(t,x,y)=\mathbf{v}_1(t,x,y_1),\ \ p(t,x,y)=p_1(t,x,y_1).\end{aligned}\tag{28}$$

As a result, we may re-write equations (9)-(13) in the form (the index 1 for the independent variables is omitted):

$$\frac{\partial\theta}{\partial t}-\frac{\partial\theta}{\partial y_1}\frac{\partial\zeta}{\partial t}+\mathbf{v}\bullet\widetilde{\nabla}\theta=\widetilde{\Delta}\theta+W_Z(\theta)\phi(\alpha),\tag{29}$$

$$\frac{\partial\alpha}{\partial t}-\frac{\partial\alpha}{\partial y_1}\frac{\partial\zeta}{\partial t}+\mathbf{v}\bullet\widetilde{\nabla}\alpha=L_e^{-1}\widetilde{\Delta}\alpha+W_Z(\theta)\phi(\alpha),\tag{30}$$

$$\mathbf{v}+\widetilde{\nabla}p=R_p(\theta+\theta_0)\gamma,\tag{31}$$

$$\frac{\partial v_x}{\partial x}-\frac{\partial v_x}{\partial y_1}\frac{\partial\zeta}{\partial x}+\frac{\partial v_y}{\partial y_1}=0.\tag{32}$$

Here $\widetilde{\Delta}=\frac{\partial^2}{\partial x^2}+\frac{\partial^2}{\partial y_1^2}-2\frac{\partial\zeta}{\partial x}\frac{\partial^2}{\partial x\partial y_1}+\left(\frac{\partial\zeta}{\partial x}\right)^2\frac{\partial^2}{\partial y_1^2}-\frac{\partial^2\zeta}{\partial x^2}\frac{\partial}{\partial y_1}$, and $\widetilde{\nabla}=\left(\frac{\partial}{\partial x}-\frac{\partial\zeta}{\partial x}\frac{\partial}{\partial y_1},\frac{\partial}{\partial y_1}\right)$.

In order to obtain the matching conditions across the reaction zone, we introduce for the inner solution the stretched coordinate $\eta = y_1/\epsilon$. Then, we look for the inner solution in the form of expansion

$$\begin{aligned}&\theta=\epsilon\tilde{\theta}^1+\ldots,\ \ \alpha=\epsilon\tilde{\alpha}^1+\ldots,\\ &\mathbf{v}=\tilde{\mathbf{v}}^0+\epsilon\tilde{\mathbf{v}}^1+\ldots,\ \ p=\tilde{p}^0+\epsilon\tilde{p}^1+\ldots,\ \ \zeta=\tilde{\zeta}^0+\epsilon\tilde{\zeta}^1+\ldots.\end{aligned}\tag{33}$$

If we substitute these expansions into (29)-(32), we obtain:

<u>Order ϵ^{-1}</u>

$$\left(1+\left(\frac{\partial\tilde{\zeta}^0}{\partial x}\right)^2\right)\frac{\partial^2\tilde{\theta}^1}{\partial\eta^2}+\exp\left(\frac{\tilde{\theta}^1}{1+\delta\tilde{\theta}^1}\right)\phi(\tilde{\alpha}^1)=0,\tag{34}$$

$$\left(1+\left(\frac{\partial\tilde{\zeta}^0}{\partial x}\right)^2\right)\frac{\partial^2\tilde{\alpha}^1}{\partial\eta^2}+\exp\left(\frac{\tilde{\theta}^1}{1+\delta\tilde{\theta}^1}\right)\phi(\tilde{\alpha}^1)=0,\tag{35}$$

$$-\frac{\partial\tilde{p}^0}{\partial\eta}\frac{\partial\tilde{\zeta}^0}{\partial x}=0,\ \ \frac{\partial\tilde{p}^0}{\partial\eta}=0,\tag{36}$$

$$-\frac{\partial\tilde{v}_x^0}{\partial\eta}\frac{\partial\tilde{\zeta}^0}{\partial x}+\frac{\partial\tilde{v}_y^0}{\partial\eta}=0.\tag{37}$$

<u>Order ϵ^0</u>

$$\tilde{v}_x^0+\frac{\partial\tilde{p}^0}{\partial x}-\frac{\partial\tilde{\zeta}^0}{\partial x}\frac{\partial\tilde{p}^1}{\partial\eta}-\frac{\partial\tilde{\zeta}^1}{\partial x}\frac{\partial\tilde{p}^0}{\partial\eta}=0,\tag{38}$$

$$\tilde{v}_y^0+\frac{\partial\tilde{p}^1}{\partial\eta}=-R_p\theta_0,\tag{39}$$

$$\frac{\partial\tilde{v}_x^0}{\partial x}-\frac{\partial\tilde{\zeta}^0}{\partial x}\frac{\partial\tilde{v}_x^1}{\partial\eta}-\frac{\partial\tilde{\zeta}^1}{\partial x}\frac{\partial\tilde{v}_x^0}{\partial\eta}+\frac{\partial\tilde{v}_y^1}{\partial\eta}=0.\tag{40}$$

Let consider both the outer and the inner expansions of the temperature for example (the same technique can be used to find the expansions of α and $\mathbf{v}$) and recall that $\eta = y_1/\epsilon$, so

$$\theta(x,y_1)=\theta^0(x,y_1)+\epsilon\theta^1(x,y_1)+\epsilon^2\theta^2(x,y_1)+\ldots,$$
$$\theta(x,\epsilon\eta)=\epsilon\tilde{\theta}^1(x,\epsilon\eta)+\epsilon^2\tilde{\theta}^2(x,\epsilon\eta)+\ldots$$

In determining the outer and the inner expansions, we use two different limit processes as adopted in [17, 18] :
The outer limit $\theta^0(x,y_1)=\lim\limits_{y_1\,fixed,\,\epsilon\to0}\theta(x,y_1)$ and the inner limit $0=\lim\limits_{\eta fixed\epsilon\to0}\theta(x,y_1)$.
Then, we use the following matching principle:

$$\lim_{y_1\to\pm0}\theta^0(x,y_1)=\lim_{\eta\to\pm\infty}0=0,$$

which corresponds to the behavior of the basic solution at $y_1 = 0\pm$ (see (8)).

Now, we write the outer solution in terms of the inner variable η, we use the Taylor expansion, the outer and the inner limit process again,

$$\frac{\partial\theta^0}{\partial\eta}(x,0)\eta+\theta^1(x,0)=\lim_{y_1 fixed \epsilon\to 0}\frac{\theta(x,y_1)-\theta^0(x,0)}{\epsilon},$$

$$\tilde{\theta}^1(x,\eta)=\lim_{\eta\, fixed,\epsilon\to 0}\frac{\theta(x,\epsilon\eta)}{\epsilon},$$

since $\frac{\partial\theta^0}{\partial\eta}(x,0)=0$ when $y_1\to 0-$, the following matching conditions hold:

$$\eta\to+\infty:\ \tilde{\theta}^1\sim\theta^1|_{y_1=0+}+\eta\frac{\partial\theta^0}{\partial y_1}|_{y_1=0+},\ \tilde{\alpha}^1\to 0,\ \tilde{\mathbf{v}}^0\to\mathbf{v}^0|_{y_1=0+},\tag{41}$$

$$\eta\to-\infty:\ \tilde{\theta}^1\to\theta^1|_{y_1=0-},\ \tilde{\alpha}^1\sim\alpha^1|_{y_1=0-}+\eta\frac{\partial\alpha^0}{\partial y_1}|_{y_1=0-},\ \tilde{\mathbf{v}}^0\to\mathbf{v}^0|_{y_1=0-}.\tag{42}$$

From (36) we remark that $\tilde{p}^0$ does not depend on η, this implies that at the leading order the pressure is continuous through the interface. From (38) and (39) we can easily deduce that $\tilde{v}^0_x$ and $\tilde{v}^0_y$ do not depend on η, this provides the continuity of the velocity across the interface. We next derive the jump conditions for the temperature from (34), in the same way as it is usually done for combustion problems. As we have a zero-order reaction, we have $\phi(\tilde{\alpha}^1)\equiv 1$. From (34), we conclude that $\frac{\partial^2\tilde{\theta}^1}{\partial\eta^2}\le 0$, so $\frac{\partial\tilde{\theta}^1}{\partial\eta}$ is decreasing from $\eta=-\infty$ to $\eta=+\infty$.
From (42) we have, $\frac{\partial\tilde{\theta}^1}{\partial\eta}\sim 0$ at $\eta=-\infty$, then $\frac{\partial\tilde{\theta}^1}{\partial\eta}\le 0$ everywhere, this shows that $\tilde{\theta}^1$ is decreasing too.

Thus multiplying (34) by $\frac{\partial\tilde{\theta}^1}{\partial\eta}$ and integrating, we obtain

$$\Big(\frac{\partial\tilde{\theta}^1}{\partial\eta}\Big)^2\Big|_{\eta=+\infty}-\Big(\frac{\partial\tilde{\theta}^1}{\partial\eta}\Big)^2\Big|_{\eta=-\infty}=\frac{2}{A}\int_{-\infty}^{\theta^1|_{y_1=0+}}\exp\Big(\frac{\tau}{1+\delta\tau}\Big)d\tau,\tag{43}$$

where we have set $A=1+\Big(\frac{\partial\tilde{\zeta}^0}{\partial x}\Big)^2$.

Using now the matching conditions and truncating the expansions:

$$\theta^0\approx\theta,\ \theta^1|_{y_1=0-}\approx Z\theta|_{y_1=0}\ ,\zeta^0\approx\zeta,\ \mathbf{v}\approx\mathbf{v}^0,\tag{44}$$

we obtain the jump conditions

$$\Big(\frac{\partial\theta}{\partial y_1}\Big)^2\Big|_{y_1=0+}-\Big(\frac{\partial\theta}{\partial y_1}\Big)^2\Big|_{y_1=0-}=2Z\Big(1+\Big(\frac{\partial\zeta}{\partial x}\Big)^2\Big)^{-1}\int_{-\infty}^{\theta|_{y_1=0}}\exp\Big(\frac{\tau}{Z^{-1}+\delta\tau}\Big)d\tau.\tag{45}$$

Since $\delta=\frac{R_0T_b}{E}$ with a large activation energy E, δ is a small parameter, we can write (34)-(35) as

$$A\frac{\partial^2\tilde{\theta}^1}{\partial\eta^2}+exp(\tilde{\theta}^1)=0,\tag{46}$$

$$A\frac{\partial^2\tilde{\alpha}^1}{\partial\eta^2}+exp(\tilde{\theta}^1)=0.\tag{47}$$

If we substrate equation (47) from (46), since $A\neq 0$, we have

$$\frac{\partial^2}{\partial\eta^2}(\tilde{\theta}^1-\tilde{\alpha}^1)=0.\tag{48}$$

By integrating twice equation (48) with respect to η and by using the above matching conditions we obtain easily:

$$\tilde{\theta}^1-\tilde{\alpha}^1=\theta^1|_{y_1=0-},\tag{49}$$

and

$$\left[\frac{\partial \tilde{\theta}^0}{\partial y_1}\right] - \left[\frac{\partial \tilde{\alpha}^0}{\partial y_1}\right] = 0, \ [\tilde{\theta}^0] = [\tilde{\alpha}^0] = 0. \tag{50}$$

Eliminating $\tilde{\theta}^1$ between equations (47) and (49) we find

$$v\frac{\partial v}{\partial u} = \Lambda e^{-u}, \tag{51}$$

where $u = \tilde{\alpha}^1$, $v = \dfrac{\partial u}{\partial \eta}$, $\Lambda = \dfrac{\exp(\theta^1|_{y_1=0-})}{A}$, with the boundary conditions $u = 0, \ v = 0, \ u = -\infty, \ v = \dfrac{\partial \alpha^0}{\partial y_1}\Big|_{y_1=0-}$.

Integrating equation (51), the following jump condition held

$$\left[\frac{\partial \alpha^0}{\partial y_1}\right]_{(y_1=0)} = \frac{\sqrt{2}}{\sqrt{A}}\exp(\theta^1|_{y_1=0-}), \tag{52}$$

when $y_1 < 0, \ \alpha^1 = 0$ then $S(y_{1-0}, x, t) = \theta^1(y_{1-0}, x, t)$ and the jump condition (52) can be written as

$$\left[\frac{\partial \alpha^0}{\partial y_1}\right]_{(y_1=0)} = \frac{\sqrt{2}}{\sqrt{A}}\exp(S|_{y_1=0-}). \tag{53}$$

Now we are able to seek a new jump condition which can be obtained from the analysis of the terms of the order ϵ^2 of the equations (14) and (27).
Indeed we have

$$\frac{\partial^2}{\partial \eta^2}(\tilde{\theta}^2 - \tilde{\alpha}^2 + l_e\tilde{\alpha}^1) = 0, \tag{54}$$

by integrating we obtain

$$\frac{\partial}{\partial \eta}(\tilde{\theta}^2 - \tilde{\alpha}^2 + l_e\tilde{\alpha}^1) = Q(x,t), \tag{55}$$

where $Q(x,t)$ is an unknown function, by connecting the inner and the outer values of the function:

$$\frac{\partial}{\partial y_1}(\theta - \alpha),$$

$$\left[\frac{\partial}{\partial y_1}(\theta^1 - \alpha^1)\right]_{y_1=0} = \lim_{\eta\to+\infty}\frac{\partial}{\partial \eta}(\tilde{\theta}^2 - \tilde{\alpha}^2) - \lim_{\eta\to-\infty}\frac{\partial}{\partial \eta}(\tilde{\theta}^2 - \tilde{\alpha}^2),$$

by using (54) we obtain the new jump condition

$$\left[\frac{\partial S}{\partial y_1}\right]_{(y_1=0)} = l_e\left[\frac{\partial \alpha^0}{\partial y_1}\right]_{(y_1=0)}. \tag{56}$$

4. Formulation of the interface problem

Here we present a complete formulation of the free boundary problem derived above.
We have for $y \neq y_1$ (in the unreacted medium):

$$\frac{\partial \theta}{\partial t} + \mathbf{v}\bullet\nabla\theta = \Delta\theta, \tag{57}$$

$$\frac{\partial \alpha}{\partial t} + \mathbf{v}\bullet\nabla\alpha = \Delta\alpha, \tag{58}$$

$$\frac{\partial S}{\partial t} + \mathbf{v}\bullet\nabla S = \Delta(S + l_e\alpha), \tag{59}$$

$$\mathbf{v} + \nabla p = R_p(\theta + \theta_0)\gamma, \tag{60}$$

$$\nabla \bullet \mathbf{v} = 0. \tag{61}$$

We complete this system by the following jump conditions at the interface $y = y_1$:

$$[\theta] = 0,\ [\alpha] = 0,\ [S] = 0,\ \left[\frac{\partial \theta}{\partial y}\right] - \left[\frac{\partial \alpha}{\partial y}\right] = 0,\ [\mathbf{v}] = [\mathbf{v}'] = 0, \tag{62}$$

$$\left[\left(\frac{\partial \theta}{\partial y}\right)^2\right] = -\frac{2Z}{1 + (\frac{\partial \zeta}{\partial x})^2} \int_{-\infty}^{\theta(x,\zeta)} \exp\left(\frac{s}{1/Z + \delta s}\right) ds, \tag{63}$$

$$\left[\frac{\partial S}{\partial y}\right] = l_e \left[\frac{\partial \alpha}{\partial y}\right],\ \left[\frac{\partial \alpha}{\partial y}\right] = \frac{\sqrt{2}\exp(S|_{y=-0})}{\sqrt{1 + \left(\frac{\partial \zeta}{\partial x}\right)^2}}. \tag{64}$$

Here [ ] denotes the jump at the interface, $[f] = f|_{\zeta-0} - f|_{\zeta+0}$. The free boundary problem is completed by the conditions at infinity:

$$y \to (+\infty, -\infty)\ \ (\theta, \alpha, S, \mathbf{v}) = ((-1,0,0,0),(0,1,0,0),(0,0,0,0),(0,0,0,0)). \tag{65}$$

5. Linear stability analysis

In this section, we perform the linear stability analysis of the steady-state solution for the interface problem that has a travelling wave solution $(\theta, \alpha, \mathbf{v})$,

$$\theta(t,x,y) = \theta_s(y - ut),\ \ \alpha(t,x,y) = \alpha_s(y - ut),\ \ S(t,x,y) = 0 \text{ and } \mathbf{v}(t,x,y) = 0, \tag{66}$$

where

$$\theta_s(t,y) = \begin{cases} 0 & \text{if } y < 0 \\ e^{-uy} - 1 & \text{if } y > 0 \end{cases}, \tag{67}$$

$$\alpha_s(t,y) = \begin{cases} 1 & \text{if } y < 0 \\ 0 & \text{if } y > 0 \end{cases}. \tag{68}$$

Here the number u stands for the stationary front velocity that can be easily found from the jump conditions at the interface.

We now introduce the moving coordinate frame $y_1 = y - ut$, for which the travelling wave is a stationary solution of the problem

$$\frac{\partial \theta}{\partial t} - u\frac{\partial \theta}{\partial y_1} + \mathbf{v} \bullet \nabla \theta = \Delta \theta, \tag{69}$$

$$\frac{\partial \alpha}{\partial t} - u\frac{\partial \alpha}{\partial y_1} + \mathbf{v} \bullet \nabla \alpha = \Delta \alpha, \tag{70}$$

$$\frac{\partial S}{\partial t} - u\frac{\partial S}{\partial y_1} + \mathbf{v} \bullet \nabla S = \Delta(S + l_e \alpha), \tag{71}$$

$$\mathbf{v} + \nabla p = R_p(\theta + \theta_0)\gamma, \tag{72}$$

$$\nabla \bullet \mathbf{v} = 0, \tag{73}$$

again with the jump conditions (62)-(64).

5.1. Perturbation problem

We now introduce a small perturbation of the reaction front to this stationary solution in the form of

$$\zeta(t,x) = ut + \xi(t,x), \tag{74}$$

$$\begin{aligned} &\theta(t,x,y) = \theta_s(y-ut) + \theta_j(z)e^{\omega t + ikx}, j = 1,2,\\ &\alpha(t,x,y) = \alpha_s(y-ut) + \alpha_j(z)e^{\omega t + ikx}, j = 1,2,\\ &S(t,x,y) = S_j(z)e^{\omega t + ikx}, j = 1,2,\\ &v_y(t,x,y) = v_j(z)e^{\omega t + ikx}, \end{aligned} \tag{75}$$

where

$$z = y_1 - \zeta(t,x) = y - ut - \xi(t,x), \;\; \xi(t,x) = \epsilon_1 e^{\omega t + ikx}, \tag{76}$$

$j = 1$ corresponds to $z < 0$ and $j = 2$ to $z > 0$.

We linearize system (69)-(73) about the stationary solution and apply *rot rot* transformation to eliminate the pressure in the equation (72). We find the following system
for $z < 0$:

$$\theta_1'' + u\theta_1' - (\omega + k^2)\theta_1 = 0, \tag{77}$$

$$\alpha_1'' + u\alpha_1' - (\omega + k^2)\alpha_1 = 0, \tag{78}$$

$$S_1'' + uS_1' - (\omega + k^2)S_1 = l_e k^2 \alpha_1 - l_e \alpha_1'', \tag{79}$$

$$v_1'' - k^2 v_1 = -R_p k^2 \theta_1, \tag{80}$$

for $z > 0$:

$$\theta_2'' + u\theta_2' - (\omega + k^2)\theta_2 + v_2 u e^{-uz} = 0, \tag{81}$$

$$\alpha_2'' + u\alpha_2' - (\omega + k^2)\alpha_2 = 0, \tag{82}$$

$$S_2'' + uS_2' - (\omega + k^2)S_1 = l_e k^2 \alpha_2 - l_e \alpha_2'', \tag{83}$$

$$v_2'' - k^2 v_2 = -R_p k^2 \theta_2. \tag{84}$$

5.2. Linearization of jump conditions

In order to linearize the jump conditions, we use the Taylor formula at the position of the reaction zone y_1 for the functions θ , α, S and v,

$$f(y_1 + \zeta \pm 0) = f_s(y_1 \pm 0) + \zeta f_s^{'}(y_1 \pm 0) + \widetilde{f}(y_1 \pm 0),$$

$$\frac{\partial f}{\partial z}\Big(y_1 + \zeta \pm 0\Big) = f_s^{'}(y_1 \pm 0) + \zeta f_s^{''}(y_1 \pm 0) + \frac{\partial \widetilde{f}}{\partial z}\Big(y_1 \pm 0\Big),$$

where f can take θ , α, S and v. For the highest order the jump conditions become: $z = y_1$

$$\theta_2(0) - \theta_1(0) = u\epsilon_1, \tag{85}$$

$$\alpha_2(0) - \alpha_1(0) = 0, \tag{86}$$

$$S_2(0) - S_1(0) = 0, \tag{87}$$

$$\theta_2'(0) - \theta_1'(0) = -\epsilon_1 u^2 + \alpha_2'(0) - \alpha_1'(0), \tag{88}$$

$$-u(u^2\epsilon_1 + \theta_2'(0)) = Z\theta_1(0), \tag{89}$$

$$S_2'(0) - S_1'(0) = l_e(\alpha_2'(0) - \alpha_1'(0)), \tag{90}$$

$$\alpha_2'(0) - \alpha_1'(0) = \sqrt{2}S_1(0), \tag{91}$$

$$v_1(0) = v_2(0) \text{ and } v_1'(0) = v_2'(0). \tag{92}$$

5.3. Solution of the linearized problem, dispersion relation

By introducing the linear differential operators $L_1\theta = \theta'' + u\theta' - (\omega + k^2)\theta$, $L_2 v = v'' - k^2 v$, equations (77)-(84) can be written as:

$$L_1\theta_1 = 0, \quad L_2 v_1 = -R_p k^2\theta_1, \tag{93}$$

$$L_1\theta_2 = -ue^{-uz}v_2, \quad L_2 v_2 = -R_p k^2\theta_2, \tag{94}$$

$$L_1\alpha_i = 0, \quad L_1 S_i = l_e k^2\alpha_i - l_e\alpha_i'', \; i = 1,2, \tag{95}$$

since the perturbations decay at infinity, so that for $i = 1,2$, we have $\theta_i(\pm\infty) = 0$, $\alpha_i(\pm\infty) = 0$, $S_i(\pm\infty) = 0$, $v_i(\pm\infty) = 0$, the general solution of system (93) has the form

$$v_1(z) = a_1\frac{R_p k^2}{k^2 - \mu_1^2}\tilde{w}_1(z) + a_2\tilde{w}_2(z), \; \theta_1(z) = a_1\tilde{w}_1(z), \tag{96}$$

where a_1 and a_2 are arbitrary constants and

$$\tilde{w}_1(z) = e^{\mu_1 z}, \; \tilde{w}_2(z) = e^{kz} \text{ and } \mu_1 = \frac{-u + \sqrt{u^2 + 4(\omega + k^2)}}{2}. \tag{97}$$

The solution of system (94) has the form

$$v_2(z) = b_1 w_1(z) + b_2 w_2(z), \; \theta_2(z) = b_1 s_1(z) + b_2 s_2(z), \tag{98}$$

where b_1 and b_2 are arbitrary constants, the functions w_i and s_i have the following form for $i = 1,2$,

$$w_i(z) = \sum_{j=1}^{\infty} a_{i,j}e^{\sigma_{i,j}z}, \; s_i(z) = \sum_{j=1}^{\infty} c_{i,j}e^{\sigma_{i,j}z}, \tag{99}$$

with

$$\begin{aligned}
&\sigma_{1,1} = \frac{-u-\sqrt{u^2+4(\omega+k^2)}}{2}, \ \sigma_{2,1} = -k,\\
&\sigma_{i,j+1} = \sigma_{i,j} - u,\\
&c_{1,1} = 1, \ c_{2,1} = 0, \ a_{2,1} = 1,\\
&a_{i,j} = \frac{R_p k^2 c_{i,j}}{k^2 - \sigma_{i,j}^2} \text{ for } (i,j) \neq (2,1),\\
&c_{i,j+1} = \frac{-u a_{i,j}}{\sigma_{i,j+1}^2 + u\sigma_{i,j+1} - (\omega+k^2)}.
\end{aligned} \tag{100}$$

The general solution of system (95) has the form

$$S_1(z) = \big(d_1 + z c_1 \frac{l_e(k^2-\mu_1^2)}{u+2\mu_1}\big)\tilde{w}_1(z), \ \ \alpha_1(z) = c_1 \tilde{w}_1(z), \tag{101}$$

$$S_2(z) = \big(d_2 + z c_2 \frac{l_e(k^2-\mu_2^2)}{u+2\mu_2}\big)\hat{w}_2(z), \ \ \alpha_2(z) = c_2 \hat{w}_2(z), \tag{102}$$

where c_1, c_2, d_1 and d_2 are arbitrary constants,

$$\hat{w}_2(z) = e^{\mu_2 z} \ \text{ and } \mu_2 = \frac{-u-\sqrt{u^2+4(\omega+k^2)}}{2}. \tag{103}$$

The constants a_1, a_2, b_1, b_2, c_1, c_2, d_1, d_2 and ϵ_1, should be found from the jump conditions (85)-(92).
Simple computations lead us to a linear system that can be written as $AX = 0$, where $X = (a_1, a_2, b_1, b_2, c_1, d_1, \epsilon_1)^T$ is the column vector, the components of which are unknown, with $c_1 = c_2$ and $d_1 = d_2$, and the matrix

$$A = \begin{pmatrix}
-1 & 0 & s_1(0) & s_2(0) & 0 & 0 & -u\\
-\mu_1 & 0 & s_1^{'}(0) & s_2^{'}(0) & -G & 0 & u^2\\
\frac{Z}{u} & 0 & s_1^{''}(0) & s_2^{''}(0) & 0 & 0 & u^2\\
0 & 0 & 0 & 0 & l_e(E-F-G) & G & 0\\
0 & 0 & 0 & 0 & G & -\sqrt{2} & 0\\
-\frac{R_p k^2}{k^2-\mu_1^2} & -1 & w_1(0) & w_2(0) & 0 & 0 & 0\\
-\frac{R_p k^2}{k^2-\mu_1^2}\mu_1 & -k & w_1^{'}(0) & w_2^{'}(0) & 0 & 0 & 0
\end{pmatrix}.$$

Here $E = \frac{k^2-\mu_2^2}{u+2\mu_2}$, $F = \frac{k^2-\mu_1^2}{u+2\mu_1}$ and $G = \mu_2 - \mu_1$.
If $det(A) = 0$ then the problem of perturbation admits non trivial solution. The solvability condition of this linear system allows us to find the dispersion relation.

5.4. Cellular stability

Basically, there are two types of instabilities: the oscillatory instability and the cellular one. 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. This corresponds to the case where $\omega = 0$. Thus, for fixed R_p, Z and u, we can draw l_e as a function of the wavenumber k for different values of the velocity u (Fig. 2). The graph shows the stable and the unstable critical areas. We remark that it doesn't depend on Z. However, we remark that when we increase the velocity u of the stationary front, it becomes less (resp. more) stable before (resp. after) some critical value of the wavenumber k_c.

Limiting case $L_e^{-1} = 0$

The variation of the critical Rayleigh number R_p depending on the wave-number k (Fig. 3) is obtained as a consequence of the dispersion relation found in the limiting case where $L_e^{-1} = 0$, as studied in [19]. In this case, the cellular instability boundaries do not depend on the Zelodovich number Z. Indeed all coefficients of Z in the dispersion relation vanish. The curve separates the stable and the unstable regions. We see that for $R_p < 0$, which corresponds to descending front, the front is always stable. Moreover, when we increase the velocity u of the stationary front, it becomes less stable [13].

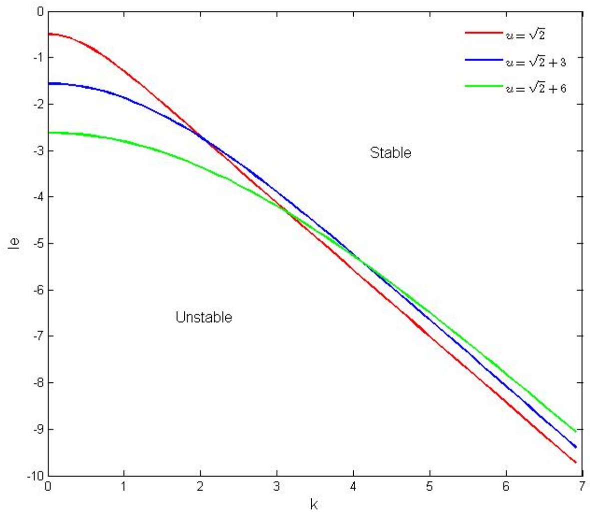

Fig. 2. Cellular stability boundary for $u = 1.41$, $u = 4.41$ and $u = 7.41$.

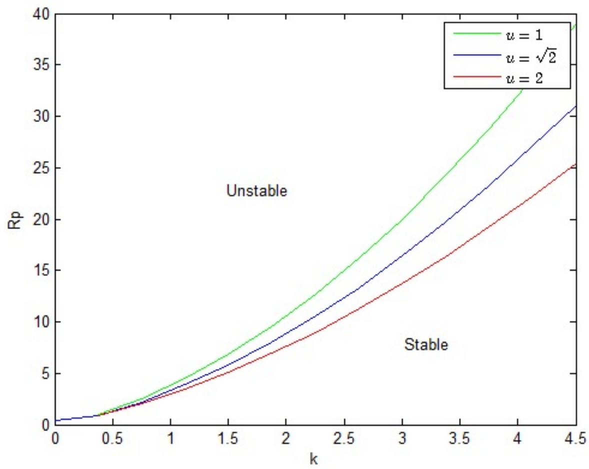

Fig. 3. Cellular stability boundary in the limiting case

6. Conclusion

In this work, the effect of the Lewis number on the convective instability of ascending polymerization fronts in porous media is studied. We first focus on the formal asymptotic analysis using the Zeldovich - Frank-Kamenetskii method. Then, from de linear stability analysis, the problem is linearized and the dispersion relation is derived in terms of the governing parameters including Zeldovich, Rayleigh and Lewis numbers, the velocity and the wave-number. The results obtained in the case of the cellular stability, show that the stability boundaries can be influenced by the Lewis number and the Rayleigh number variations.

It would also be interesting to study the oscillatory instability which corresponds to $\omega = i\psi$ where $\psi \neq 0$. In this case, we obtain a complex valued equation from the dispersion relation, that is very complicated and should be analysed numerically.

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