CONCENTRATION-DEPENDENT CONVECTION IN A HORIZONTAL POROUS BED CONTAINING A CHEMICALLY ACTIVE LIQUID

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The limits to convective stability of a mixture in a horizontal porous bed have been derived for several types of boundary conditions subject to the occurrence of a homogeneous isothermal reaction.

1. Consider a horizontal layer of porous medium filled with a chemically active binary liquid mixture, which consists of a heavy reagent and a lighter product. The latter is produced throughout the volume of the layer without change in temperature at a rate proportional to the concentration of the reagent (first-order reaction). Reactions of this type are common in various electrochemical processes. The boundaries of the layer are isothermal planes, so the temperature is constant and the density is dependent only on the concentration. The concentration variations during the reaction may result in unstable stratification and therefore in concentration-dependent convection. The appropriate conditions have been defined approximately [1] for the case where the upper boundary of the layer has a constant reagent concentration, while the latter becomes zero at the bottom. Here a more general formulation is employed.

The porous bed has thickness d and the z axis is directed vertically upwards, while the xy plane is coincident with the lower boundary of the layer. It is assumed that the linear law $\rho = \rho_0(1 + \beta C)$ applies for the density of the mixture as a function of reagent concentration.

The equations for convection are as follows if we assume Darcy's law and the usual Boussinesq approximations:

$$\begin{gathered} \frac{1}{\rho_0}\nabla p + \frac{\nu}{K}\mathbf{v} + g\beta C\mathbf{e} = 0, \\ \frac{\partial C}{\partial t} + \frac{1}{m}\mathbf{v}\nabla C = D\Delta C - kC, \\ \operatorname{div}\mathbf{v} = 0. \end{gathered} \tag{1.1}$$

The reagent concentration is constant at the upper boundary:

$$C = C^{(0)} \quad \text{at} \quad z = d. \tag{1.2}$$

Boundary conditions of several types may occur at the lower boundary $z = 0$ [2]:

a) reagent absent:

$$C = 0; \tag{1.3}$$

b) reagent flux of a given density:

$$\frac{\partial C}{\partial z} = \alpha, \tag{1.4}$$

where the case $\alpha > 0$ corresponds to withdrawal of reagent and $\alpha < 0$ corresponds to supply, while $\alpha = 0$ represents a boundary impermeable to the reagent, in which case (1.4) coincides with the condition used in [1]; and

c) a heterogeneously catalyzed first-order reaction at $z = 0$ or mass transfer at a rate proportional to the concentration:

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$$D\frac{\partial C}{\partial z} = \sigma C. \tag{1.5}$$

The normal component of the infiltration rate is zero at both boundaries for all types of condition:

$$v_z = 0 \text{ at } z = 0;\ d. \tag{1.6}$$

We write the equations of (1.1)-(1.6) in dimensionless form while retaining the previous symbols and using the quantities d, mD/d, $C^{(0)}\sqrt{D}/k/d$, $\rho_0 \nu Dm/K$, and d^2/D as the units of measurement for distance, velocity, concentration, pressure, and time, respectively; system (1.1) then becomes as follows for the dimensionless variables:

$$\begin{gathered} \nabla p + \mathbf{v} + RC\mathbf{e} = 0, \\ \frac{\partial C}{\partial t} + \mathbf{v}\nabla C = \Delta C - \Psi^2 C, \\ \operatorname{div}\mathbf{v} = 0. \end{gathered} \tag{1.7}$$

The similarity parameters are $R = g\beta KC^{(0)}/\nu m\sqrt{kD}$ (the infiltration analog of the Rayleigh number) and $\Psi = d\sqrt{k/D}$ (the Thiele modulus, which characterizes the extent of reaction). The dimensionless boundary conditions are as follows:

$$\begin{gathered} \text{at } z = 1\colon v_z = 0,\ C = \Psi; \text{ at } z = 0\colon v_z = 0; \\ \text{a) } C = 0,\ \text{b) } \frac{\partial C}{\partial z} = \alpha,\ \text{c) } \frac{\partial C}{\partial z} = \gamma C, \end{gathered} \tag{1.8}$$

where $\gamma = \sigma d/D$ is the Sherwood number.

2. The infiltration rate is zero at mechanical equilibrium and all the quantities are independent of time. A necessary condition for such equilibrium is [3] a vertical density gradient, which here is dependent on z on account of the reaction occurring in the volume. The steady-state distributions for C_0 (the concentration of the heavy component) may be determined by solving (1.7) with $\mathbf{v}_0 = 0$ and $d/dt = 0$, and these are described by the following expressions as appropriate to the types of boundary conditions:

$$\text{a) } C_0 = \Psi \operatorname{sh} \Psi z / \operatorname{sh} \Psi, \tag{2.1}$$

$$\text{b) } C_0 = (\Psi \operatorname{ch} \Psi z / \operatorname{ch} \Psi) + \alpha (\operatorname{sh} \Psi (z - 1)/\Psi \operatorname{ch} \Psi), \tag{2.2}$$

$$\text{c) } C_0 = \Psi (\Psi \operatorname{ch} \Psi z + \gamma \operatorname{sh} \Psi z)/(\Psi \operatorname{ch} \Psi + \gamma \operatorname{sh} \Psi). \tag{2.3}$$

Figure 1 shows the distributions of (2.1)-(2.3).

If there is no reagent at the lower boundary, viz., condition (1.8a) applies, then the concentration pattern is described by (2.1); the density distribution is unstable for all nonzero Ψ. The stratification of the mixture increases with the Thiele modulus, and the zone of fall in the reagent concentration becomes narrower and tends to a thin boundary layer at the $z = 1$ plane for Ψ large.

The regions of positive α in the distribution of (2.2) denote outflowing reagent, and these have an upper bound of $\alpha_+ = \Psi^2/\operatorname{sh}\Psi$, which is the condition for complete absence of the outgoing component at $z = 0$. In the limiting case ($\alpha = \alpha_+$), (2.2) reduces to (2.1). An increase in the rate of input of the reagent (an increase in the modulus for negative α) causes

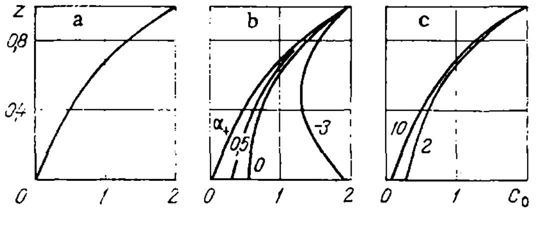

Fig. 1. Equilibrium distributions for the reagent concentration C_0 for $\Psi = 2$ corresponding to the following distributions: a) (2.1); b) (2.2) for $\alpha = -3$, 0, 0.5, α_+; c) (2.3) for $\gamma = 2$, 10.

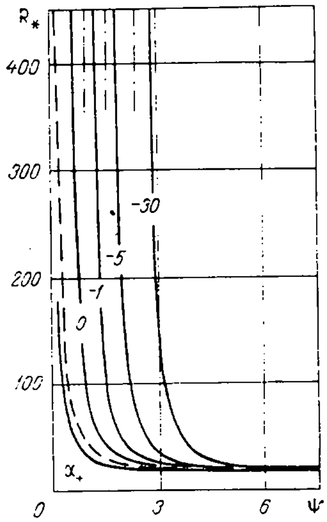

Fig. 2. Minimum critical value of the Rayleigh number R_* as a function of Ψ; the broken curves correspond to the steady-state distribution of (2.1), while the solid lines are those corresponding to (2.2) for $\alpha = \alpha_+$, 0, −1, −5, −30.

the mixture at the lower boundary to become enriched in the heavy component, which may suppress the unstable density stratification and therefore may eliminate the conditions necessary for convection to occur. Stable density distributions occur in the presence of reagents supply if $\alpha \le \alpha_- = -\Psi^2 \mathrm{sh}\Psi$, and for $\alpha = \alpha_-$ we have $dC_0/dz = 0$ at the upper boundary of the layer. The equilibrium concentration distribution of (2.2) is the same as that of [1] for $\alpha = 0$.

Boundary condition (1.8c) results in a potentially unstable stationary distribution in (2.3) for which $\Psi \neq 0$, which lies between the results of (2.1) and (2.2) for $\alpha = 0$ and coincides with these for values of γ of 0 and ∞, respectively (infinitely slow and infinitely rapid heterogeneous reactions at $z = 0$, respectively).

The equilibrium distributions of (2.2) and (2.3) approximate to the distribution of (2.1) for $\Psi \to \infty$.

3. The convective instability in these equilibrium distributions may be examined by reference to small normal perturbations in the rate and concentration that are proportional to $\exp[-\lambda t + i(a_1 x + a_2 y)]$.

We introduce the perturbations and eliminate the pressure in (1.7) to get a system of homogeneous ordinary linear differential equations for the amplitudes $w(z)$ and $\eta(z)$ of the perturbations:

$$\begin{gathered}(w'' - a^2 w) = a^2 R\eta,\\ -\lambda\eta + wC_0' = (\eta'' - a^2\eta) - \Psi^2\eta\end{gathered} \tag{3.1}$$

(a prime denotes differentiation with respect to z).

The boundary conditions for w and η are given by (1.8) as

$$\begin{gathered}\text{at } z = 1\text{: } w = 0,\ \eta = 0;\quad \text{at } z = 0\text{: } w = 0;\\ \text{a) } \eta = 0,\ \text{b) } \eta' = 0,\ \text{c) } \eta' = \gamma\eta.\end{gathered} \tag{3.2}$$

The perturbation decrements λ are eigenvalues for (3.1)-(3.2) and are derived by specifying that a nontrivial solution to (3.1) exists. The eigenvalues may be derived numerically by the Runge–Kutta–Merson method [4], which allows one to check the error of integration.

Calculations show that perturbations with real λ set the lower bound to instability and that the oscillatory situations that occur for the conditions of (1.8b) and (3.2b) are stable.

The eigenvalues become $\lambda = 0$ for certain values of the parameters in (3.1)-(3.2), and these define the stability limits corresponding to the neutral $R(a, \Psi)$ curves.

We now discuss the stability results.

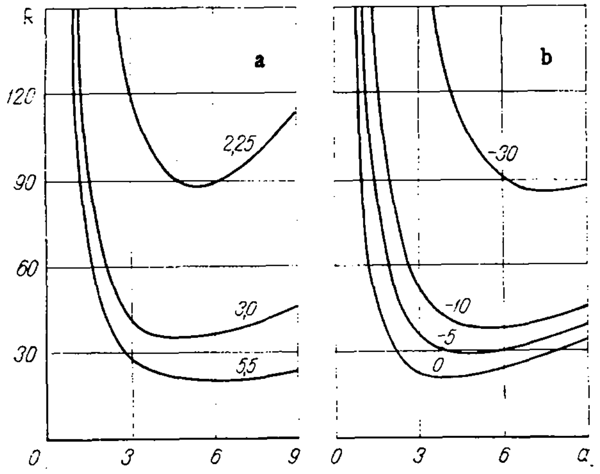

Fig. 3. Neutral-stability curves: a) $\alpha =$ -5 and $\Psi = 2.25$, 3.0, 5.5; b) $\Psi = 3.25$ and $\alpha = 0$, -5, -10, -30.

The broken line in Fig. 2 shows the minimum critical value for the Rayleigh number R_* as a function of the Thiele modulus Ψ for the absence of reagent at the lower boundary, viz., conditions (1.8a) and (3.2a); the zone of instability lies above the curve. The gradient in the equilibrium concentration is small if Ψ is small, and the corresponding critical values of R_* are large. The stratification increases with Ψ and the heavy component is located mainly in the upper part of the layer. Consequently, the $C_0(z, \Psi)$ relation on the one hand becomes more favorable to convection when Ψ is large, and the critical R_* fall, while on the other the motion occurs in a narrow region near the $z = 1$ boundary. These opposing factors result in the $R_*(\Psi)$ curve approaching the asymptote $R_* \approx 19.9$ for $\Psi \to \infty$.

Input or withdrawal of reagent at the lower boundary has a very marked effect on the convective stability of an equilibrium horizontal layer, in accordance with (1.8b) and (3.2b). Figure 2 shows the corresponding $R_*(\Psi)$ curves for various values of α (solid lines). An increase in the rate of loss of the heavy component, i.e., variation in α over the range $0-\alpha_+$, is accompanied by an increase in the density gradient and a reduction in the convective stability. The resulting family of critical characteristics $R_*(\Psi)$ lies between the curves $\alpha = \alpha_+$ and $\alpha = 0$ in Fig. 2 [note that the differences in the boundary conditions for η result in $R_*(\Psi)$ curves that do not coincide although the steady-state concentration distributions are identical with and without loss of reagent at $z = 0$].

If the heavy reagent is supplied through the lower boundary ($\alpha < 0$), the zone of unstable stratification is reduced, and the stability of the equilibrium becomes greater. Figure 2 shows $R_*(\Psi)$ curves for input at various rates ($\alpha = -1, -5, -30$). The density distributions are stable for $\alpha \leq \alpha_-$, so the $R_*(\Psi)$ curves show asymptotic behavior for certain Ψ (the vertical dot-dash lines in Fig. 2). The following transcendental equation defines the asymptotes:

$$\Psi^2 \operatorname{sh} \Psi + \alpha = 0. \qquad (3.3)$$

All Ψ less than the values given by (3.3) result in absolutely stable equilibrium. The convective stability of the system falls as the Thiele modulus increases, and R_* approaches $R_* = 19.9$ for any α, the latter result being the one derived from the boundary conditions of (1.8a) and (3.2a).

If there is no flow of reagent at the lower boundary ($\alpha = 0$), then the boundary-value problem of (3.1)-(3.2b) is the same as that considered in [1]. An approximate solution was derived in [1] by Galerkin's method by means of certain basis functions that differ as between large and small values of the Thiele modulus. The appreciable error in those results and the minimum on the $R_*(\Psi)$ curve arise from the use of a small basis along with the assumption that the system is equivalent to a semi-infinite one for Ψ comparatively small. The curve for $\alpha = 0$ in Fig. 2 represents the revised $R_*(\Psi)$ relation.

Figure 3 shows examples of neutral-stability $R(\alpha)$ curves for various Ψ for $\alpha = -5$ (Fig. 3a) and for various reagent input rates for $\Psi = 3.25$ (Fig. 3b).

The latter shows that the critical values of the wave number a_* become larger as α decreases, which means that perturbations of smaller scale can cause instability, which itself is due to reduced thickness of the region of unstable stratification.

Heterogeneous reaction or loss of reagent proportional to the concentration at the lower boundary, as represented by (1.8c) and (3.2c), will have relatively little effect on the convective stability. The family of $R_*(\Psi)$ curves for values of the Sherwood number between 0 and ∞ lies in the range between the $R_*(\Psi)$ curves corresponding to the boundary conditions of (1.8a), (3.2a) and (1.8b), (3.2b) for $\alpha = 0$.

NOTATION

v, infiltration rate; C, C_0, $C^{(0)}$, current reagent concentration, equilibrium value, and dimensional value at upper boundary; p, convective correction to pressure; t, time; g, acceleration due to gravity; e, vertical unit vector; $\beta = (1/\rho_0)(\partial\rho/\partial C)_{T,p}$, coefficient relating density to concentration; K, permeability; m, porosity; ν, D, kinematic viscosity and diffusion constant; k, rate constant of homogeneous reaction; σ, rate constant for heterogeneous reaction and mass transfer; w(z), $\eta(z)$, amplitudes of normal velocity and concentration perturbations; a_1, a_2, wave numbers for perturbations along x and y axes, $a_1^2 + a_2^2 = a^2$; λ, perturbation decrement.

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THREE-DIMENSIONAL POTENTIALS FOR THE TELEGRAPHERS' EQUATION AND THEIR APPLICATION TO BOUNDARY-VALUE HEAT-CONDUCTION PROBLEMS

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Three-dimensional potentials for the telegraphers' equation are introduced and used to reduce boundary-value heat-conduction problems to integrodifferential equations of the second kind.

In recent years the hyperbolic heat-conduction equation has been used to solve various kinds of heat-conduction and thermoelasticity problems [1, 2]. Therefore, it has become necessary to create the mathematical apparatus for solving direct and inverse heat-conduction problems based on the hyperbolic equation. With this in mind we generalize the potential method to the case of the telegraphers' equation.

Three-Dimensional Potentials for the Telegraphers' Equation. We consider the homogeneous telegraphers' equation with constant coefficients

$$-\frac{1}{c^2}\cdot\frac{\partial^2 u(t,\ M_0)}{\partial t^2}-\frac{1}{a}\cdot\frac{\partial u(t,\ M_0)}{\partial t}+du(t,\ M_0)+\Delta u=0 \tag{1}$$

and zero initial conditions

$$u(0,\ M_0)=\partial u(0,\ M_0)/\partial t=0 \tag{2}$$

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