1997

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Spatiotemporal behavior of convective Turing patterns in porous media

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(Received 20 December 1996; accepted 30 June 1997)

We investigate the effects of convection on Turing patterns in porous media. We show that convectionless patterns can only exist confined to small domains. These patterns are unstable to convection if the density gradients are large enough. The numerical solution of the Schnackenberg model coupled to Darcy’s law shows that the convective pattern is either steady, oscillatory, or reverses direction, depending on the density gradient. In larger domains, we find that convection leads to an oscillatory state which becomes steady for large density gradients. © 1997 American Institute of Physics. [S0021-9606(97)51837-X]

I. INTRODUCTION

Forty years after Alan Turing’s prediction, experiments verified the existence of Turing pattern formation in a chemical system. These patterns evolve from a homogeneous steady state due to the interaction between chemical species with distinctly different diffusion coefficients. These patterns are steady variations of chemical concentrations on a spatial domain. Turing proposed his reaction-diffusion model to explain biological morphogenesis, the formation of pattern and form in the embryo. The first observations took place in a chemical reactor containing a polyacrilamide gel instead of a biological system. Further experiments showed the existence of Turing patterns in a gel-free medium. The chemical gradients associated with Turing patterns may lead to density gradients which can cause convective fluid motion. Convection, in turn, may either modify or destroy the pattern. The importance of convection in chemical pattern formation is already well established. In the Beloussov–Zabotinski reaction, convection causes a significant increase in the speed of the fronts. The front is distorted as it propagates upwards in a vertical tube. In the iodate–arsenous reaction, convection is responsible for a change in front curvature and for fingering when the fluid is confined in a vertical slab (a Hele–Shaw cell). In other systems, the differential fluid velocity between chemical species is necessary for the formation of the chemical pattern. Today there are no experimental observations of the effects of convection in Turing pattern formation. Most of the experiments are carried out inside gels which prevent fluid motion. Nevertheless, convection may play an important role in Turing patterns that appear in nature, such as in biological systems. The importance of a complete model that accounts for the mechanical process was acknowledged by Alan Turing in his seminal paper on pattern formation. He acknowledged that a complete model should take into account Newton’s law of motion and other possible interactions, “But even so it is a problem of formidable mathematical complexity...The interdependence of chemical and mechanical data adds enormously to the difficulty, and attention will therefore be confined, so far as is possible, to cases where these can be separated.” It is the purpose of this work to explore the interaction between the chemical pattern and the fluid flow induced by it. This should motivate further experiments designed to observe predicted behavior.

A previous study of Turing patterns in viscous fluids showed that convection plays an important role in pattern selection since some patterns can coexist with convection while others are destroyed. A weakly nonlinear analysis showed that convection may help to stabilize some Turing patterns. There have been studies relating the differential flow mechanism of chemical pattern formation with Turing patterns. In the present work we study the role of convection in Turing patterns confined in porous media. The purpose of this study is to predict new structures that may arise from the interaction between convective fluid motion and Turing patterns. We model the flow as flow in porous media using Darcy’s law. It has the added advantage that the equations are easier to solve than the Navier–Stokes equations because Darcy’s law is of lower order. This allows us to treat patterns extending into larger domains. We will show that certain patterns are unstable to convection, and that convection caused by Turing pattern formation can lead to oscillatory behavior.

II. THE MODEL

We choose to study Turing pattern formation using the standard Schnackenberg model. This model has been studied extensively without convection. The choice of the model is not very important at this point since the fluid parameters are still unknown. We are interested in predicting new patterns that may arise from the interaction between fluid motion and the Turing mechanism. The Schnackenberg model consists of four reaction steps:

\[ k_1 A \rightarrow X, \]

\[ k_2 X \rightarrow \text{Products}, \]

\[ k_3 2X + Y \rightarrow 3X, \]

\[ k_4 B \rightarrow Y. \]

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The chemicals $X$ and $Y$ obey the mass action law while the pool chemicals $A$ and $B$ remain constant. The Schnackenberg model coupled with Darcy’s law provides the equations of motion for the chemical system:

$$\mathbf{V} = -\frac{K}{\mu} (\nabla P + \rho g \hat{z}),$$

$$\nabla \cdot \mathbf{V} = 0,$$

$$\frac{\partial X}{\partial t} + (\mathbf{V} \cdot \nabla) X = D_X \nabla^2 X + k_1 A - k_2 X + k_3 X^2 Y,$$

and

$$\frac{\partial Y}{\partial t} + (\mathbf{V} \cdot \nabla) Y = D_Y \nabla^2 Y + k_4 B - k_3 X^2 Y.$$

Here, $\mathbf{V}$ is the fluid velocity, $P$ is the pressure, $g$ is the acceleration of gravity in the vertical $z$ direction, $K$ is the coefficient of permeability of the medium, and $\mu$ is the coefficient of viscosity. The density difference is included only where it modifies the large gravity term. We assume that the density $\rho$ varies linearly with the concentration $X$:

$$\rho = \rho_0 [1 - \alpha (X - X_0)].$$

Here, $\alpha$ is the coefficient of linear expansion and $\rho_0$ is the density when $X = X_0$, the reference concentration. In this work we assume that $\alpha$ is positive and, consequently, the density is lower at higher concentrations of $X$. We neglect thermal expansion which may arise in exothermic reactions. We consider only two-dimensional movement confined in the $x-z$ plane. Since the velocity field has zero divergence, we make use of the stream function $\psi$ to eliminate the pressure from Eq. (1). The components of the fluid velocity are related to the stream function by

$$V_x = \frac{\partial \psi}{\partial z}, \quad \text{and} \quad V_z = -\frac{\partial \psi}{\partial x}.$$  

We introduce dimensionless units

$$t' = \frac{D_X t}{L^2}, \quad x' = \frac{x}{L}, \quad \frac{d}{D_X} = \frac{L^2 k_2}{D_X}, \quad \gamma = \frac{L^2 k_2}{D_X},$$

$$a = \frac{k_1}{k_2} \frac{1}{k_2} \frac{k_3}{k_2}^{1/2} A, \quad b = \frac{k_4}{k_2} \frac{k_3}{k_2}^{1/2} B,$$

$$u = \frac{k_3}{k_2}^{1/2} X, \quad v = \frac{k_3}{k_2}^{1/2} Y, \quad \mathbf{V}' = \frac{L}{D_X} \mathbf{V},$$

$$\psi' = \psi / D_X, \quad \omega' = \omega / D_X,$$

and

$$Ra = \frac{K g \alpha L \rho_0}{D_X \mu} \frac{k_2}{k_3}^{1/2}.$$  

The dimensionless parameter $Ra$ is equivalent to the Rayleigh number defined for thermal convection in porous media. The length $L$ is the typical length scale of the system and will be defined later. With these substitutions, the set of nonlinear equations that govern the time evolution of the system is

$$\frac{\partial u}{\partial t} = \frac{\partial (\psi, u)}{\partial (x, z)} + \nabla^2 u + \gamma (a - u + u^2 v),$$

$$\frac{\partial v}{\partial t} = \frac{\partial (\psi, v)}{\partial (x, z)} + d \nabla^2 v + \gamma (b - u^2 v),$$

and

$$\nabla^2 \psi = -Ra \frac{\partial \psi}{\partial u}.$$

We study Turing patterns confined in rectangular boxes, therefore the boundary conditions correspond to no chemical flow and no normal velocity at the side walls. The parameters $a$, $b$, and $d$ determine the stability of the homogeneous convectionless steady state. The set of parameters that leads to Turing pattern formation has been studied extensively elsewhere. Since we are interested mainly in the effects of hydrodynamics on a well developed pattern, we set $a = 0.14$, $b = 1.1$, and $d = 20$ to obtain a convectionless Turing pattern. The convectionless Turing pattern develops with a characteristic wavelength that depends on the choice of parameters. Choosing the length scale $L$ to closely approximate half the wavelength leads to a dimensionless parameter $\gamma = 40$. Therefore, a rectangular box of dimensionless height 1 and dimensionless length 0.5 can accommodate only one type of Turing pattern that extends for half its wavelength in the vertical direction and has no structure in the horizontal direction.

### III. NUMERICAL METHODS

We solved the equations numerically using two different methods. One is a finite difference approach where we discretize the spatial domain using a rectangular mesh. The other is a truncated Fourier expansion of the chemical concentrations and the stream function. For the finite difference method, we varied the spatial grid depending on the box size. Small size boxes can only accommodate simple confined patterns while larger boxes can accommodate more complex patterns. We used a five-point expansion to approximate the Laplacian on the rectangular mesh. The time evolution was calculated using the simple Euler method with a small time step depending on the size of the mesh. A large mesh requires a smaller time step. The Poisson equation [Eq. (8)] was solved using the GENBUN routine from the FISHPACK package which makes use of a cyclic reduction technique.

We also developed a method based on a two-dimensional Fourier expansion of the chemical concentrations and the stream function. This method allowed us to model confined patterns using a few terms of the expansion. The reduced model consists of a set of ordinary differential…
equations with very few variables. Consequently, it requires less computer time. The small number of variables also allows a linear stability analysis of a simple convectionless pattern. We expanded the chemical concentrations using a Fourier expansion on a two-dimensional rectangular box of horizontal length $L_x$ and vertical height $L_z$:

$$u = \sum_{m=0}^{\infty} \sum_{n=0}^{\infty} u_{nm} \cos \left( \frac{n \pi x}{L_x} \right) \cos \left( \frac{m \pi z}{L_z} \right)$$

(10)

with a similar expansion for the variable $v$. The coefficients of the expansion are time dependent. The expansion for the stream function is of the form:

$$\psi = \sum_{m=1}^{\infty} \sum_{n=1}^{\infty} \psi_{nm} \sin \left( \frac{n \pi x}{L_x} \right) \sin \left( \frac{m \pi z}{L_z} \right)$$

(11)

These expansions match the boundary conditions at the walls: the stream function is zero and the normal derivatives of the concentrations vanish. Substituting the expansions into the set of partial differential equations and projecting over the basis functions leads to a set of ordinary differential equations (ODE) for the expansion coefficients $u_{nm}$ and $v_{nm}$. The Poisson equation [Eq. (8)] relates the coefficients for the stream function, $\psi_{nm}$, to the coefficients for the chemical variables. Thus, the resulting system of ODE’s only involves the coefficients for the chemical variables. This is the standard Galerkin expansion requiring the residue of the equations to be orthogonal to the basis functions. This method is analogous to the Lorenz method for thermally driven convection which showed a transition to chaos. Once the system was in place, it was solved with the simple Euler method.

**IV. RESULTS**

We study first the effects of convection on Turing patterns confined in boxes of dimensions $L_x \times 1$. This box allows the formation of a convectionless Turing pattern of half a wavelength in the vertical direction and no gradient in the horizontal direction. We tested the stability to convection of this pattern by carrying out a linear stability analysis on a six-term Fourier expansion per concentration variable. This implies keeping the terms in Eq. (10) with $n + m = 3$. The same order truncation of the stream function requires only three terms instead of six. The convectionless steady solution corresponds to $u_{00} = 1.24$, $u_{01} = 0.7853$, $u_{02} = 0.039$, $v_{00} = 0.70238$, $v_{01} = -0.19841$, $v_{02} = -0.0041$, and the remaining terms are equal to zero. This solution represents a pattern with the higher concentration of the chemical $u$ underneath a lower concentration of the same chemical. The vertical gradient in the concentration induces a density gradient with low density fluid underneath high density fluid, which may be unstable to convection. The reversed solution (light fluid above heavier fluid) is also valid and is obtained by changing all signs except for the variables $u_{00}$ and $v_{00}$. The linear stability analysis was carried out by linearizing the corresponding set of ordinary differential equations around the convectionless solution. The partial derivatives required for this linearization were computed numerically. The eigenvalues of the linear matrix were calculated using routines in the EISPACK diagonalization package. For a given value of $L_x$, we look for the critical Rayleigh number with the largest eigenvalue equal to zero. Values of the Rayleigh number smaller than the critical value result in negative eigenvalues, and consequently, the convectionless solution is stable. A positive eigenvalue indicates a perturbation growing exponentially with time, which corresponds to an unstable solution.

The linear stability analysis gives the variation of the critical Rayleigh number as a function of the horizontal length $L_x$ (Fig. 1). We notice that for small values of $L_x$ the critical Rayleigh number increases as $L_x$ decreases. This is due to the fact that fluid motion is more difficult in smaller domains, therefore it is harder for convection to set in. Consequently, the pattern is more stable in smaller domains. As the domain size is further increased, we observe that the Rayleigh number decreases until it reaches a minimum near $L_x = 0.83$. The existence of this minimum is similar to the case of Rayleigh–Benard convection in a confined two-dimensional box. In three dimensions this minimum does not exist, it may also disappear in our case. We observe a transition point near $L_x = 1.2$ where the slope of the curve is discontinuous. At this transition point a double zero appears in the set of eigenvalues. The transition to convection changes from a one-roll transition to a two-roll transition. The fluid flow near the onset for $L_x < 1.2$ consists of fluid rising on one side of the box and fluid descending on the other side, as shown in Fig. 2. For boxes with length $L_x > 1.2$ we find two counter-rotating convective rolls near the onset of convection (Fig. 3). These vector fields were calculated using the corresponding eigenvectors of the linear system at the critical Rayleigh number. The eigenvectors were used in the Fourier expansion to reconstruct the stream function, allowing the calculation of the vector components of the velocity. Since the eigenvectors are defined up to an ar-

![FIG. 1. The critical Rayleigh number for the onset of convection on a box of variable length and height equal to 1 in dimensionless units. In this pattern the lighter fluid is underneath the heavier fluid. There is no gradient in the horizontal direction. The convectionless pattern is stable for Rayleigh numbers below the critical Rayleigh number.](image-url)
bitrarily constant, the figures represent the shape and relative magnitudes of the velocity field.

The solutions of the nonlinear equations show a steady convective state for Rayleigh numbers just above the onset of convection. The numerical solutions of the nonlinear system were compared to the results of the linear stability analysis, using the convectionless steady state with small random perturbations as the initial state. The nonlinear evolution equations for \( L_x = 0.5 \) show that the perturbations died out for values of \( Ra < 12.8 \) and grow into a steady convective state for \( Ra \) slightly above 12.8. The results of the simulations are summarized in Fig. 4 where the dimensionless kinetic energy (defined as \( \frac{1}{2} \int |V|^2 \)) is plotted as a function of the Rayleigh number. For Rayleigh numbers below 12.8 there is no convection. For Rayleigh numbers just above this critical value we find that a single steady convective roll develops. The fluid motion is similar to the one described in Fig. 2. The kinetic energy helps to quantify the magnitude of the convective fluid motion. As the Rayleigh number increases, an oscillatory state is observed. This oscillatory state occurs for Rayleigh numbers between 27.42 and 27.84 and a small range of values. There is a small region of bistability between the oscillatory state and steady convection. As we increase the Rayleigh number beyond the oscillatory regime, the pattern reverses completely. The lighter fluid moves above the heavier fluid and convection stops. These results are different from thermal convection where a transition to complex flow (chaos) is observed when the Rayleigh number is increased. This is because the temperature gradient is imposed externally at the boundaries, while in Turing patterns the concentration gradient is imposed by the pattern itself, allowing it to reaccommodate into other forms.

The results of the Fourier expansion compare well with the results of the finite difference calculations. The box of dimensions \( 0.5 \times 1 \) was partitioned in a \( 12 \times 24 \) mesh. The time evolution of the system was carried out with the simple Euler method using a time step of \( 3.32 \times 10^{-5} \). The results are shown in Fig. 4 where they are compared with the results of the Fourier expansion model. The behavior found is similar to the one obtained previously. The convectionless pattern loses stability to convection as the Rayleigh number increases. Increasing it further leads to an oscillatory state. Finally the pattern reverses itself, stopping convection. The difference between the two methods lies in the bifurcation points. While the Fourier expansion exhibits a transition to convection at \( Ra = 12.8 \), the finite difference simulations indicate a critical Rayleigh number between 15 and 16 for the onset of convection. The oscillatory regime occurs for Rayleigh numbers between 34.1 and 34.8, which is a higher value than the corresponding regime on the Fourier expansion method. The reason for the discrepancy is that in the Fourier expansion only very few terms were employed. The truncation with few terms allowed us to carry out the linear
stability analysis and obtained the fundamental characteristics of the bifurcation sequence.

To obtain complex Turing patterns, one must consider boxes of larger dimensions, where the convectionless model allows the formation of different types of patterns. In Fig. 5 we show two stable patterns forming in a box of dimension $2 \times 1$. The pattern in Fig. 5(a) has no gradient in the horizontal direction, its stability depends on the value of the Rayleigh number. For a Rayleigh number above 5.1 the pattern is unstable, small random perturbations grow, convection develops, and eventually the lighter fluid moves above the heavier fluid stopping convection. The final result is the same pattern but reversed. The pattern in Fig. 5(b) has a gradient in the horizontal direction as well as in the vertical direction. This will force convection to be present even for small values of the Rayleigh number. The steady convectionless pattern is simply not a solution of the hydrodynamic equations. This result is displayed in Fig. 6, where we plot the kinetic energy as a function of the Rayleigh number. The kinetic energy grows as the Rayleigh number is increased. Convection distorts the pattern as the light fluid tends to rise. The fluid flow consists of two counter-rotating convective rolls at each side of the pattern. The flow is very similar to the one displayed in Fig. 7, although the dimensions of the box are different. As the Rayleigh number is increased above 10.4 we find that the pattern can no longer sustain convection. The density difference forces the lighter fluid to move completely above the heavier fluid. The pattern evolves into the first pattern with the light fluid completely on top. The final pattern is the one displayed in Fig. 5(a) but with the light fluid on top. This process shows that convection plays an important role in pattern selection, since it can carry one type of pattern into another.

Patterns in larger boxes can sustain convection for very large Rayleigh numbers. In a box of size $1.4 \times 1.4$, a convectionless pattern with gradients in the horizontal and vertical direction develops. The only other patterns allowed correspond to rotations of the initial pattern. As the Rayleigh number increases, the fluid motion increases. The kinetic energy of the fluid increases as shown in Fig. 8. In this case we did not observe a transformation into another type of convectionless pattern. We gradually increased the Rayleigh number to 200 without observing any instability of the steady convective flow. The only effect observed was an increasing distortion of the initial pattern accompanied by an increase in the kinetic energy. This effect is shown in Fig. 9 where we have compared the solutions for Rayleigh numbers of 25 and 100. The distortion caused by two convective rolls (Fig. 7) is larger for larger Rayleigh numbers.

We studied the effects on complex convective patterns using a box of dimension $5 \times 5$. Since the free pattern wave-
length (2 in dimensional spatial units) has a smaller dimension than the box size, it allows a more complex structure. The resulting convectionless pattern leads to density gradients in the horizontal and vertical directions. Regions of lighter fluid intercalate with regions of heavier fluid [Fig. 10(a)]. Convection sets in for small values of the Rayleigh number. Using the convectionless pattern as the initial condition, a relatively small Rayleigh number equal to 0.75 leads to an oscillatory convective state. This oscillation is verified by looking at the chemical concentration \( u \) on a specific location at the coordinates \( x=1.8 \) and \( z=2.4 \). The oscillations periodically fluctuate between a minimum (0.761) and maximum (2.63) value for the variable \( u \) (dimensionless units). The period of the oscillation is 3.8 (dimensionless time). As the value of the Rayleigh number is increased to 1.5, the oscillations have a smaller amplitude (0.79 for the minimum and 2.60 for the maximum) and a shorter period (3.2 time units). This trend continues as the Rayleigh number is further increased (Fig. 11). As the Rayleigh number is increased in steps of 0.75, the amplitude of the oscillations diminish. Buoyancy forces help to spread lower regions of light fluid, separating from the initial state, and creating a bubble that moves to the surface. In Fig. 10(b) we show the pattern with two of the light spots spreading near the bottom. The temporal behavior is depicted in Fig. 12. We took a vertical slice at \( x=1.8 \) in the box and observed the time evolution. This shows how the light liquid spreads and moves up. This is a periodic process, which is repeated successively. For Rayleigh numbers larger than 7.5 there are no oscillations, and the Turing patterns reaccom-

![FIG. 8. The kinetic energy as a function of Rayleigh number on a 1.4 × 1.4 box. The light fluid cannot move completely above the heavy fluid since that configuration is not allowed by the Turing mechanism. The kinetic energy will increase with increasing Rayleigh number.](image)

![FIG. 9. Convective Turing patterns in a 1.4×1.4 box. The pattern (a) has \( Ra=25 \), while (b) is for \( Ra=100 \). There is a distortion of the pattern for the higher Rayleigh number.](image)

![FIG. 10. Turing patterns in a 5×5 box. The pattern (a) has \( Ra=0 \) and has no convective fluid motion. Pattern (b) has \( Ra=3 \) and it oscillates. Pattern (c) is for \( Ra=10.5 \) and is steady. Pattern (d) is also for \( Ra=10.5 \) but developed from different initial conditions.](image)

![FIG. 11. The chemical concentration \( u \) at the coordinates (1.8,2.4) for different Rayleigh numbers. The squares represent the maximum and minimum values as the concentration oscillates. The triangles are steady values for higher Rayleigh numbers. The pattern was formed in a 5×5 box.](image)
modulate leading to a steady convective state shown in Fig. 10(c). This state consists of four elongated strips with mirror symmetry around a vertical line through the center of the box. The increase of the Rayleigh number changes the shape of the pattern, but no oscillatory or more complex temporal behavior develops. In such a box, the stable convectionless pattern is not unique. Different initial conditions provide a slightly different convectionless pattern. However, the temporal behavior has the same characteristics of the pattern described above. The pattern oscillates, the oscillations diminish with increasing Rayleigh number, and finally a steady state sets in for large Rayleigh numbers. The final state is shown in Fig. 10(d). This state also consists of elongated vertical regions similar to the previous state [Fig. 10(c)], but instead of four regions, three thicker regions appear. This difference is due to the confinement of the pattern.

V. CONCLUSIONS

We have shown that convection plays an important role in Turing pattern formation. Convectionless patterns can exist only in highly confined domains where only a vertical gradient can exist. The stability of this pattern however is determined by the vertical density gradient induced by the Turing patterns. If the density gradient is large enough, these patterns are unstable, leading to steady convection. In some cases the density difference causes the light fluid to move completely above the heavier fluid stopping convection.

As we test the effects of convection on more extended domains, we find that convection will always be present in patterns having a vertical and horizontal gradient. This convection modifies the pattern, leading to a convectionless pattern if the domain size allows one, or to very distorted patterns generated by the convective fluid flow. In these cases convection can be sustained even for very large density gradients, without the need of an external density gradient as is the case of a viscous fluid heated from below. This fact, that the density gradient is imposed internally, allows the pattern to reaccommodate into convectionless patterns or patterns with steady convection. In the Rayleigh–Bénard experiment, a transition to turbulent flow is caused by an increase in the thermal gradient. In the case of Turing patterns this is not necessarily the case. A large chemical gradient leads to a steady pattern with a steady fluid flow.

An important result of this work is the observation of an oscillatory solution induced by the density gradient. This solution was obtained for patterns that are less confined, allowing many patterns of several wavelengths inside the domain. These flow-induced oscillations may be observed in experiments. We estimated the Rayleigh number using the fractional density difference of the iodate–arsenous acid reaction equal to $10^{-4}$ as the value for its analog $\beta (k_3/k_1)^{1/2}$, the Turing pattern wavelength of 0.3 mm ($L=0.15$ mm), the coefficient of permeability as in the Hele–Shaw cell approximation $K = L^2/12$, the molecular diffusivity of water $D = 2 \times 10^{-5} \text{ cm}^2/\text{s}$, the viscosity of water $\nu = \mu / \rho_0 = 9.2 \times 10^{-3} \text{ cm}^2/\text{s}$, and the acceleration of gravity $g = 980 \text{ cm/s}^2$. With these values we obtained $Ra = 0.15$, which is relatively close to $Ra = 0.75$, where our model shows an oscillatory behavior on a $5 \times 5$ box. However, a direct comparison with experiments should also consider the experimental chemical mechanism instead of the Schnackenberg model. For large enough gradients, the pattern becomes steady showing an asymmetry between the vertical and horizontal direction. This type of pattern may be observable in experiments similar to the ones performed in a capillary tube. Turing patterns can be created in tubes of different diameters and the system can be rotated to look for distortions along the vertical direction.

ACKNOWLEDGMENTS

The authors thank Joseph Wilder for many discussions and for a critical reading of the manuscript. This research was supported by an award from Research Corporation.