A reaction–diffusion (RD) system that grows axially as one of its boundaries moves is equivalent to a boundary-forced open flow in which all species have identical flow coefficients. Depending on the flow or growth rate, \( \phi \), and on the intrinsic spreading velocity, \( c_0 \), of the RD structure, such systems are either absolutely \( (\phi < c_0) \) or convectively \( (\phi > c_0) \) unstable. We previously showed how periodic boundary forcing of an axially growing domain could be used to control the formation of space-periodic structures in biological morphogenesis. This paper proposes, as a chemical equivalent of an axially growing embryo, the design of a continuously fed unstirred flow reactor (CFUR), characterized by a photo-chemically controlled moving boundary. Using the Turing-unstable CDIMA system as an example, we illustrate by simulations the kinds of wave structures that are expected to arise in the absolutely and convectively unstable regimes when boundary forcing is either constant or time-periodic.

1. Introduction

It has been known for some time\(^{1-4}\) that the spatio-temporal dynamics of so-called *convectively unstable* open flow systems is determined by the temporal dynamics at its inflow boundary. Recent theoretical studies\(^{5-9}\) have shown how a constant boundary forcing causes space-periodic structures to form spontaneously in plug-flow reactors containing an oscillating chemical medium. The differential transport that is usually associated with the breaking of spatial symmetry\(^{10,11}\) is not required in this case and all species may have identical flow and diffusion coefficients. This phenomenon was verified experimentally\(^{12}\) and it was shown that the space-periodic structure is actually a phase wave generated as the flow distributes the temporal oscillation in space.\(^{13,14}\) Also investigated was the case of periodic boundary forcing.\(^{15-16}\) In that case, volume elements entering the flow at different times have different phases. The results are upstream or downstream travelling phase waves of constant\(^{13,14}\) or oscillatory\(^{15}\) velocity.

We recently extended our analysis of periodically forced open flows of oscillatory media to open flows of Turing-unstable and bistable media\(^{17}\) where the interacting activator and inhibitor have different diffusion coefficients but equal flow coefficients \( \phi \). In general, the flow converts a periodic boundary forcing with frequency \( \omega \) (period \( T = 2\pi/\omega \)) into a spatial mode with wave-number \( k = c_0/\phi \), (wavelength \( \lambda = \phi T \)). This mode may be amplified and maintained when the imposed wave number lies within an appropriate range supported in the absence of a flow and when the flow system is linearly or nonlinearly convectively unstable.\(^{17}\) Convectively unstable conditions arise when the flow velocity is greater than the intrinsic velocity \( c_0 \) with which a perturbation spreads in the absence of a flow. In the *absolutely unstable* regime, the mode imposed by the boundary forcing competes with the intrinsic mode as well as with modes that are excited either by noise within the system or boundary noise. The latter, rather than the boundary forcing, may determine its spatio-temporal dynamics and produce a so-called noise-sustained structure.\(^{1,2}\)

The *local control of global patterning* by the boundary forcing of convectively unstable open flow systems has key implications, for instance in biological morphogenesis. This is because any system in which a boundary moves relative to a medium, for instance the cells in an axially growing tissue, from a mathematical point of view, is equivalent to an open flow where all species have identical flow coefficients.\(^{15,14,17}\) This equivalence is easy to verify. Consider a boundary that moves through a stationary medium with velocity \( -\phi \). Now change to the reference frame where the boundary remains stationary. In this reference frame the medium moves away from the boundary, i.e. downstream, at a constant velocity \( +\phi \), as it would in an open flow system where all the flow coefficients are equal. Whether it is the medium or the boundary that actually moves is irrelevant, as long as there is a relative motion between them. This relative motion between the medium and its boundary is what resolves a time-periodic boundary forcing into a space-periodic mode.

This paper investigates the control of pattern formation by a moving boundary of illumination in the light-sensitive\(^{20,21}\) chlorine dioxide iodide malonic acid (CDIMA) reaction\(^{18,19}\) in a continuously fed unstirred reactor (CFUR).\(^{22,23}\) We suggest how an experimental investigation of patterns that are controlled at a moving boundary may be carried out. The dynamics of the CDIMA reaction is known to be controllable by illumination with visible light. According to the current mechanism,\(^{20,21}\) iodine atoms produced by photo-dissociation of molecular iodine initiate reduction of chlorine dioxide and
oxidation of iodide ions to iodine. As a result, oscillations and formation of Turing structures may be conveniently suppressed by a sufficiently intense illumination.

We envisage an experimental setup as shown in Fig. 1a. The CFUR is rectangular with lengths $L_x$ and $L_y$. Two masks block the light in the left-most region of the CFUR while the right side is illuminated sufficiently intensely to suppress formation of Turing structures. Mask 1 is stationary and allows one to control the left-most boundary condition when mask 2 has not advanced beyond it. In what follows, laterally uniform or non-uniform symmetries will be imposed onto the left-hand boundary. Mask 2 is moved with velocity $\phi$ from left to right on top of mask 1 until it covers the entire reactor. As a result the non-illuminated, Turing-active portion of the reactor grows with velocity $\phi$.

The moving mask establishes the moving illumination boundary illustrated in Fig. 1b. When it is transversely uniform, the light-intensity, $w(x,y,t)$, within the CFUR is given by:

\[ w(x,y,t) = w_0(t)H(x), \]

where $H(x_0)$ is the Heavyside function: $H(x) = 1$ if $x > x_0$ and $H(x) = 0$ if $x < x_0$ for all $y$ in the reactor domain. Note that $w(x,y,t)$ is expressed in dimensionless units (see eqns. (2)–(3) below). As illustrated in Fig. 1b, eqn. (1) describes a moving boundary, located at $x = x_0$, between two regions of different illumination. One region is in the shade ($H(x) = 0$ if $x < x_0$) while the other is illuminated with intensity $w(x) = w_0(t)$.

The boundary of illumination moves with a constant velocity $\phi$ and its position is given by $x = x_0 + \phi t$, where $x_0$ is the initial, $t = 0$, location of the boundary. The shape of mask 1 allows for initial conditions of different lateral symmetries. We consider here either a flat, laterally uniform mask or the T-shaped mask illustrated in Fig. 1a.

The CDIMA reaction illuminated by light with space- and time-dependent intensity $w(x,y,t)$, can be described by the two-dimensional reaction–diffusion system:20,21

\[ \frac{\partial u}{\partial t} = \alpha - \beta u - \frac{4w}{1 + w^2} - w(x,y,t) + D_u \left( \frac{\partial^2 u}{\partial x^2} + \frac{\partial^2 u}{\partial y^2} \right), \]

\[ \frac{\partial v}{\partial t} = \sigma \left( \beta u - \frac{vw}{1 + w^2} + w(x,y,t) \right) + \sigma D_v \left( \frac{\partial^2 v}{\partial x^2} + \frac{\partial^2 v}{\partial y^2} \right), \]

where $u$ and $v$ are dimensionless concentrations of the activator ($I^-$) and the inhibitor ($\text{ClO}_2^-$); $\alpha$, $\beta$, and $\sigma$ are dimensionless parameters proportional to other initial concentrations and rate constants. $D_u$ and $D_v$ are the diffusion coefficients of activator and inhibitor. Simulations were performed using a finite-difference scheme and no-flux boundary conditions at $x = 0$ and $x(t) = \phi = t = L_y$ as well as at the lateral boundaries $y = 0$, $L_x$.

To begin our discussion, we investigate in section 2 the linear stability of the uniform steady state of the CDIMA system in eqns. (2)–(3) in the one-dimensional case without a moving boundary. This analysis provides the modes of the non-illuminated system that are amplified when a perturbation with a real wave number $k$ is applied. Subsequently, the effects of spatially uniform, constant and periodic illumination are studied. Section 3 explores the effects of the moving illumination boundary under absolutely unstable ($\phi < \phi_c$) and under convectively unstable ($\phi > \phi_c$) conditions in one and two spatial dimensions.

### 2. Turing-structures in the CDIMA reaction

It is well known that in the absence of illumination the CDIMA reaction system may sustain spatial non-uniform patterns when subject to perturbations with wave numbers within a certain range.18,21 The photosensitive CDIMA system in eqns. (2)–(3) has the following uniform steady state $S = (u_s,v_s)$ given by:

\[ S = \left( \frac{x - 5w}{5\beta}, \frac{2(x - 5w)^2 + 25\beta^2}{25\beta(x - 5w)} \right) \]

(4)

This steady state exists only in the parameter domain: $x > 5w$, $w \geq 0$ and $\beta > 0$. The temporal stability of $S$ to spatial perturbations is determined by the linearized equations around $S$. If the geometry is assumed one-dimensional (i.e. small $L_x$) the calculations give the eigenvalues associated with wave number $k$ of a small amplitude spatial perturbation around the homogeneous steady state. Perturbations with an eigenvalue that has a positive real part grow in time while those with negative real part decay. Fig. 2 show the bifurcation diagram for the stability regions of the system in eqns. (2)–(3) for $w = 3$ and $\beta = 0.3$ in the $(x,\sigma)$ plane of the kinetic parameters. The Hopf and Turing boundaries are shown. The point $P$ of coordinates $x = 16$, $\sigma = 20$, where the illumination suppresses the Turing pattern, will be used in the numerical investigations below.
Fig. 3a shows the biggest eigenvalue as a function of the wave number (squared) in the absence of illumination, \( w = 0 \). The modes with wave numbers between \( k_1^2 = 0.23 \) and \( k_2^2 = 0.56 \) (wavelengths between 8.3 and 12.9) are unstable. Spatially random small amplitude perturbations having wave numbers in this range are amplified and may develop into large amplitude Turing patterns.

The effect of constant illumination on the range of unstable wave numbers is shown in Fig. 3b. For low intensities, i.e. for \( w < 2.6 \), illumination destabilizes modes that are stable when \( w = 0 \). The destabilizing effect of the light is most pronounced for short wavelengths (high values of \( k \)). The range of unstable wave numbers is maximal at about \( w = 2.55 \) where modes between \( k_1^2 = 0.19 \) and \( k_2^2 = 3.0 \) (wavelengths between 3.6 and 114.4) are unstable. Turing structures are suppressed at high light intensity. With the parameter values used in Fig. 2, all modes are stable when \( w \) exceeds 2.82.

Fig. 4 illustrates the suppression of Turing patterns in the illuminated region of the CFUR. The boundary between the illuminated and the non-illuminated regions is kept fixed at \( x_0 = L_x/2 = 100 \). The left half of the space-time plot in Fig. 4a shows the development of a stable Turing structure in the non-illuminated region. The large concentration gradients established at the boundary between the two regions causes a Turing structure to spread into the non-illuminated region at a velocity \( c_0 \) that is slightly greater than unity. Eventually, it fills the entire non-illuminated region. In contrast, the illuminated region remains homogenous. Fig. 4b shows the stable concentration profiles of \( u \) and \( v \) when all transients have died out.

It was recently demonstrated\(^{22,23} \) that periodic illumination could also be used to suppress the formation of Turing patterns. In that case, the periodic illumination is taken to be:

\[
\nonumber w_0(t) = \frac{w_a}{2} \left( 1 + \sin \left( \frac{2\pi t}{T} \right) \right).
\]

where \( T \) is the period of the illumination cycle and \( w_a \) is its amplitude. The suppression of Turing structures by periodic illumination is illustrated in Fig. 5. At constant illumination, spatial homogeneity is only achieved when the light intensity lies above a critical value.
3. Moving boundary of illumination

We now turn to the case where the boundary between the illuminated and the non-illuminated region moves at a constant velocity. As mentioned in the Introduction, the system may be either convectively unstable (at high $\phi$) or absolutely unstable (at low $\phi$). Fig. 6 illustrates the effect of having a moving boundary of illumination and constant light intensity, $w_0 = 3$. This high-intensity light suppresses the Turing patterns in the region ahead of the moving boundary.

The boundary moves at a relatively low velocity, $\phi = 1$, in Fig. 6a and at a relatively high velocity, $\phi = 3$, in Fig. 6b. In Fig. 6a, a Turing-structure fills the entire non-illuminated region at all times. This is because the Turing-structure spreads at a velocity $c_T$ that is greater than the velocity of the moving boundary (absolutely unstable flow conditions). For the case in Fig. 6b, the boundary moves faster than the Turing structure can spread so the region immediately behind the moving boundary remains homogenous. This corresponds to convectively unstable flow conditions. Absolutely and convectively unstable systems have different properties. The two cases are discussed in separate sub-sections below.

3.1 Absolutely unstable conditions

A key feature of absolutely unstable flow conditions is that the final pattern depends on the initial condition. This can be used to select between different transverse symmetries by changing the shape of mask 1 in Fig. 1a. Fig. 7a and b shows the stripe and, respectively, the hexagonal pattern that is selected at constant illumination when the mask is transversely uniform (Fig. 6a) or when it is T-shaped (Fig. 6b).

In contrast to the case of convectively unstable conditions discussed in section 3.2 below, the absolutely unstable system is rather insensitive to periodic illumination ahead of the moving boundary. Fig. 7c shows the effect of having periodic illumination, eqn. (5), with a period $T$ and amplitude $w_a = 3$. Although the combination of the moving boundary ($\phi = 1$) and the periodic forcing imposes a transversely uniform mode (wavelength $\phi T = 12$) it is the hexagon-pattern, selected by the initial condition that prevails.

3.2 Convectively unstable conditions

In marked contrast to the absolute unstable system, the convectively unstable system is rather insensitive to its initial conditions. This is because the boundary moves at a velocity $\phi$ that is greater than the velocity $c_T$ at which the Turing structure spreads. Hence the region immediately behind the moving boundary remains uniform (see Fig. 6b) regardless of the initial condition.

While the absolutely unstable system is insensitive to periodic forcing ahead of the moving boundary, the convectively unstable system is very sensitive to such manipulation. As described in the Introduction, the relative movement between the medium and the boundary causes temporal modes ahead of the boundary to be converted into spatial modes behind it. The spatial modes may be amplified if they have wave numbers $k = \omega/\phi$ (wavelength $\lambda = \phi T$) in the appropriate, unstable range (Fig. 3). This is illustrated in Fig. 8.

The four space-time plots presented in Fig. 8 are obtained with different periods of illumination as described in eqn. (5). In Fig. 8a, the periodic illumination ($T = 2$) and the movement of the boundary ($\phi = 3$) imposes a spatial mode with a wavelength of $\lambda = 6$. This mode has eigenvalues with negative real parts (see Fig. 2) and the region behind the moving boundary is homogenous.

In Fig. 8b ($T = 3$) and Fig. 8c ($T = 4$) the mode imposed onto the medium by the movement of the boundary has an eigenvalue with a positive real part (see Fig. 2). As a result, the mode is amplified and a stable Turing structure develops in the region behind the moving boundary. As mentioned in the Introduction, the velocity of the boundary and the illumination period determines the wavelength of the spatial mode imposed by the boundary forcing. It is therefore possible to control the particular wavelength of the resulting pattern by choosing appropriate values for $\phi$ and $T$.

Fig. 9 illustrates the control of pattern formation in a two-dimensional system by periodic illumination in the convectively unstable system. The initial condition, seen in the top frame, is established in the same way as in Fig. 7b and c. The flow velocity is $\phi = 3$ and the illumination period is $T = 4$. The wavelength of the imposed mode is $\lambda = 12$. This is the same wavelength as that imposed to the absolutely unstable system in Fig. 7c. Recall from Fig. 7c that the periodic illumination had little effect on the hexagonal pattern selected due to the transversely non-uniform initial condition.
Integration time is 100 time units. In contrast, it is the transversely uniform stripes that prevail in the convectively unstable system shown in Fig. 9.

4. Discussion

We have presented a numerical study that suggests how Turing patterns in the CDIMA reaction can be controlled at an axially moving boundary of illumination. The crucial control parameter is the speed of the moving shadow mask.

When the velocity of the moving boundary is sufficiently low and the system is absolutely unstable, it is possible to favour a certain lateral symmetry by choosing the appropriate symmetry of the boundary condition. A transversely uniform boundary condition favours a Turing structure composed of transversely uniform stripes. On the other hand, a T-shaped boundary condition with the appropriate dimensions favours a hexagonal Turing structure.

When the boundary moves with a velocity that is sufficiently high to make the system convectively unstable, then the initial lateral symmetry is less important. Since noise is inevitably present at the moving boundary, and noise tends to be amplified by convectively unstable systems, a noise-sustained structure is expected to arise. It is therefore unlikely that the spatially uniform region behind the moving boundary in Figs. 6b, 8a and 8d can be observed experimentally. However, by applying a periodic illumination ahead of the moving boundary, i.e. time-periodic boundary conditions, it should be experimentally feasible to impose transversely uniform stripes. This is not a trivial result since periodic illumination does not correspond to the sinusoidal low amplitude perturbation considered in the linear analysis (section 2) and in our earlier investigations that established the phenomenon. The wavelength of the resulting Turing pattern depends on the velocity of the moving boundary and on the illumination period, both of which are experimentally adjustable. It should therefore be possible to control not only the symmetry of the Turing pattern but also to select its wavelength. This may in turn be used to probe the range of unstable wave numbers and their growth rates.

Finally, the proposed experimental setup is a chemical equivalent of developing biological systems where the movement of the boundary results from a reorganization of the tissue or from cell divisions within a terminal growth zone.

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References